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**A Study of Vegetable Sourced Colourants  
for use in  
Screen Process Ink Production**

A thesis submitted to Middlesex University  
in partial fulfillment of the requirements for the degree of  
Doctor of Philosophy

Philip T. Shaw

School of Art, Design and the Performing Arts  
Visual Communication Design

Middlesex University

2000



## ACKNOWLEDGEMENTS

In 1994 I was fortunate enough to obtain research funding for a project which was eventually to lead to the following study. Were it not for the faith placed in my project by Middlesex University's School of Art, Design and Performing Arts Research Team and their continuing support over the past six years, the present work would not have been possible. I owe all those who gave that support a debt of gratitude.

In particular, I would like to note my special appreciation for the help given to me by Professor Barry Jackson, Gill Hewitt, Steve Mumberson, Professor Brian Shutes and Dr. Ian Williams, their vigilance and good advice in all aspects of the work were of immense value. I would also like to thank Dr. Rob Withnall of Greenwich University, Dr. John Davison of Davison Chemographics, Dr. Dominique Cardon, Dr. David Cooke and Dr. Kerry Stoker of Bristol University, Colin Thurston, Jenny Dean of the Ashill Colour Studio, Keith Howard of the Canadian School for Non-toxic Printmaking, Henryk Bøegh, Emma Karp and all my students round the world for much practical assistance and encouragement. My thanks must also be expressed to my colleagues in Visual Communication Design for their help, encouragement and interest. My two children, Florence and Arthur must also be credited for some stirring work behind the camera - my love and appreciation to them and finally, to Susan Light for her affectionate support and practical assistance throughout the often difficult stages of the work.

## **Abstract**

The following investigation aims to determine the potential of plant material to produce pigments capable of acting as alternatives to the standard range of process (CMYK) colours used in screen printing.

A review of the literature on so-called 'natural dyes' reveals the extent of the pre-synthetic (1857) use of a range of dye-plants in the textile and related industries over a period of many centuries. Several plant sources are identified for investigation in the context of this study. Chemical, botanical and historical aspects of these plants are described along with notes on their cultivation and nomenclature. The necessary thickeners and other additives that may be required for the production of a stable ink formulation are also discussed.

The establishment of the *Cat Hill Ink Garden* is described as well as a discussion of other sources of raw material supply. Methods of aqueous extraction, lake production, comparative light fastness and spectral reflectance testing are all described and test results are provided in appendix form.

The environmental, economic and health and safety aspects of the new inks is contrasted with the commercial oil-based product as well as a brief discussion of their potential for commercial development.

Printed examples by the author and several of his students form the final part of the study and demonstrate the use of both CMYK and other non-primary ink formulations.

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## CHAPTER I

### Introduction

It is almost a century since the introduction of screen printing, a technique which adapted and refined the very ancient principle of stenciling into a commercially viable reprographic process. Throughout the 1930's and particularly after 1945 the process was developed to a point where it began to rival lithographic printing particularly in the field of poster work. Throughout its development, the industry, in common with lithographic and letterpress printing, used predominantly oil-based inks with pigmentation from mineral or synthetic sources. The introduction over the past decade of water-based screen inks, has reduced and in some cases eliminated the need for volatile organic solvents, providing a much safer environment for workers in the industry. The pigmentation of these water-based inks however, remained and continues to remain, essentially 'non-renewable' being largely synthesised from mined mineral compounds, coal tar and petroleum products. Over the past decade, a considerable effort has been made by industry to address the concerns of environmental and government agencies regarding sustainability, the environment and health and safety. My initial thoughts at the outset of this investigation concerned the more mundane problem of increased expenditure and diminishing resources within the university printmaking department - could we make our own inks more cheaply

than we could buy them? This initial question led to the eventual realisation that it may be possible to develop a truly renewable source of colour, which could be utilised in printing inks and would present a practical alternative to the materials currently available.

### The Aims of this Study

The aim of this study is to investigate the possibility of using hitherto neglected vegetable colours in screen process printing inks and in particular to try to find equivalents for the so-called 'process' or 'CMYK' colours used in the reproduction of photographic images on paper and card. This proposed application is wholly novel. The historical use of vegetable-based 'block printing' colours in Japan and elsewhere, as well as the experiments carried out by William Morris in the late 19th century, pre-date the introduction of both screen printing and the modern 'four colour' process. No attempts at the preparation of 'process' colours from plant material for use in screen printing inks, is recorded in the literature.

### Research Questions

The research questions addressed in this study are:

- What vegetable sources are capable of providing suitable colours in

the context of the research aims?

- How might these colours compare chromatically with standard process (CMYK) colours?
- How do they compare with existing colours in terms of light fastness?
- What methods and materials can be employed in their formulation as screen printing inks?
- What advantages might be derived from the use of such colours?

### Background to the Study

The substances within all plants that provide their startlingly varied colours are said to be of three types, anthocyanins, anthoxanthins and chlorophyll. The water soluble anthoxanthins are responsible for many of the yellow pigments which abound in plant material, and are the chief constituents of traditional dyes like weld, buckthorn and fustic (see Chapter II). The anthocyanins, also soluble in water, are found largely in flowers and fruits, producing the reds, blues and purple pigments therein - cornflowers, poppies and elderberries being examples of plants which have been variously used to make pigments and dyes of this type. Whilst the anthocyanin pigments can produce vivid reds and purples when processed into



dyestuffs, they have been found for the most part to be rather fugitive to light though the application of certain metallic salts are said by some to produce more durable colours .

Chlorophyll, the green pigment present in the cells of leaves, responsible for absorbing light energy for photosynthesis, has rarely been used as a dye or pigment due largely to its instability in light. It is used today however to colour a variety of food products including ice cream, chewing gum and soups (Hanssen, 1987), where its lack of fastness to light is not so important. In Europe it is designated as the natural food additive E 140, and listed in the Colour Index as Natural Green 3 (CI 75810).

By the early part of the 20th century a considerable body of research into the chemistry of plant pigments had accumulated and workers like A.G. Perkin and A.E. Everest, Rawson, Gardner and Laycock and later J. Mayer had all published seminal works which are still regarded as works of authoritative reference. While a basic understanding of chemistry has been necessary in order to progress my investigations, it is not my purpose to discuss chemistry in detail. My background and training is in the practical and fine arts and detailed reference to the chemistry of plant material would require a degree of expertise that I simply do not possess. Rather it has been my

policy to introduce only as much technical discussion as is necessary to convey an understanding of my argument. My approach has been throughout to investigate practical possibilities and to offer the results of those investigations to others who may be able to bring to bear more specialised scientific, technical and commercial expertise, where further development seems appropriate.

The plant derived colours investigated in this study, whose main aim is to find equivalents for the four, so-called 'process' colours used in full colour print reproduction, has focused mainly on the yellows obtained from weld (*Reseda luteola*) and buckthorn (*Rhamnus sp.*). The reds from madder (*Rubia tinctoria*), elder (*Sambucus nigra*), pokeweed (*Phytolacca americana* syn. *P. decandra*) and brazilwood (*Caesalpinia echinata*). Blue, being the most difficult colour to achieve using plant material will be obtained largely from the indigo bearing species which include indigo itself (*Indigofera tinctoria*) but which can also be derived from Japanese knotweed (*Polygonum tinctoria*), woad (*Isatis tinctoria*) and others. Blacks will be derived from various combinations, including logwood (*Haematoxylon campechianum*), elder (*Sambucus nigra*), walnut hulls, and a variety of barks containing high concentrations of hydrolysable tannin eg. oak and oak galls, alder, sumac etc. The necessary vegetable

hydrolysable tannin eg. oak and oak galls, alder, sumac etc. The necessary vegetable thickeners, preservatives and notes on cultivation will also be discussed.

### The Four 'Process' Colours

In order to print a full colour reproduction of any given image, using just four colours (yellow, magenta, cyan and black), the original artwork, or a photographic version of it, is separated into these four constituent colours and processed onto separate transparent film positives, where they appear as fine dot patterns, large dots being the most intense colour areas, small dots giving the impression of paler hues. This is achieved today using laser scanning technology, in the past, coloured photographic filters were used to achieve the same end. Green, purple, orange and brown areas, being the result of various combinations of the four process colours will consequently appear on more than one of the film positives. The positive representing black provides the tonal range from dark to light. These four positives can then become separate printing plates, as in the case of lithography, or for screen process printing as stencils supported by a fine silk, synthetic or stainless steel mesh.

Because the scanning procedure results in four internationally

recognised standard colour separations, special standard inks must be used if a faithful reproduction of the original is to be achieved.

Trichromatic, or process inks, as we know them today, have been manufactured now for some fifty years, before which time a rather unsatisfactory blue meant that colour reproduction was less than ideal. The development of Phthalocyanine blue in the ICI laboratories during the 1930's provided the print industry with what is essentially the range used today. Diarylide and quinacridone yellows and reds are typical sources of process pigments each, in common with phthalocyanine blue being entirely synthetic products derived from non-renewable resources.

In order to reproduce in print full colour work using vegetable sourced pigments, equivalent, or near equivalents of the process colours currently in use must be found. This is the essential problem which this study proposes to address. An idea of the difficulties involved can be seen in the spectral reflectance curves shown in Figs. 1, 2 and 3 taken from one of my initial colour separation test prints. Along the way, a range of colours, unsuitable for use as process colours will inevitably be developed, and these will not be ignored or discarded, since they may form a separate range of standard printing inks.

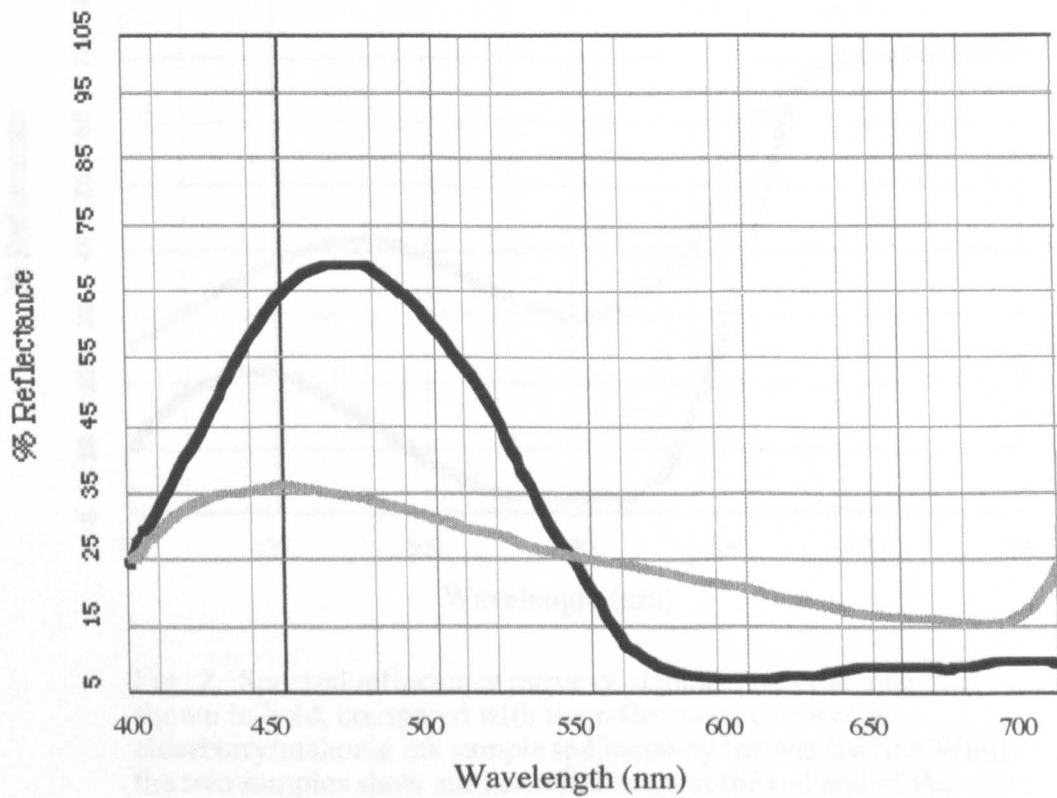


Fig. 1. Spectral reflectance curve of standard process cyan shown in bold, compared with the reflectance curve of an indigo print sample, indicated by the shaded line. In the standard sample the peak around 470 nm. is that area of the spectrum responsible for blue/green, or cyan reflectance. It can be readily seen that the indigo sample has only a very modest peak in that area, along with a pronounced peak around 700 nm. the area responsible for red reflectance. In simple terms, we can determine from this that the colour being represented is a fairly dull blue, which also contains a touch of red - in other words indigo.

Courtesy of Davison Chemographics Ltd

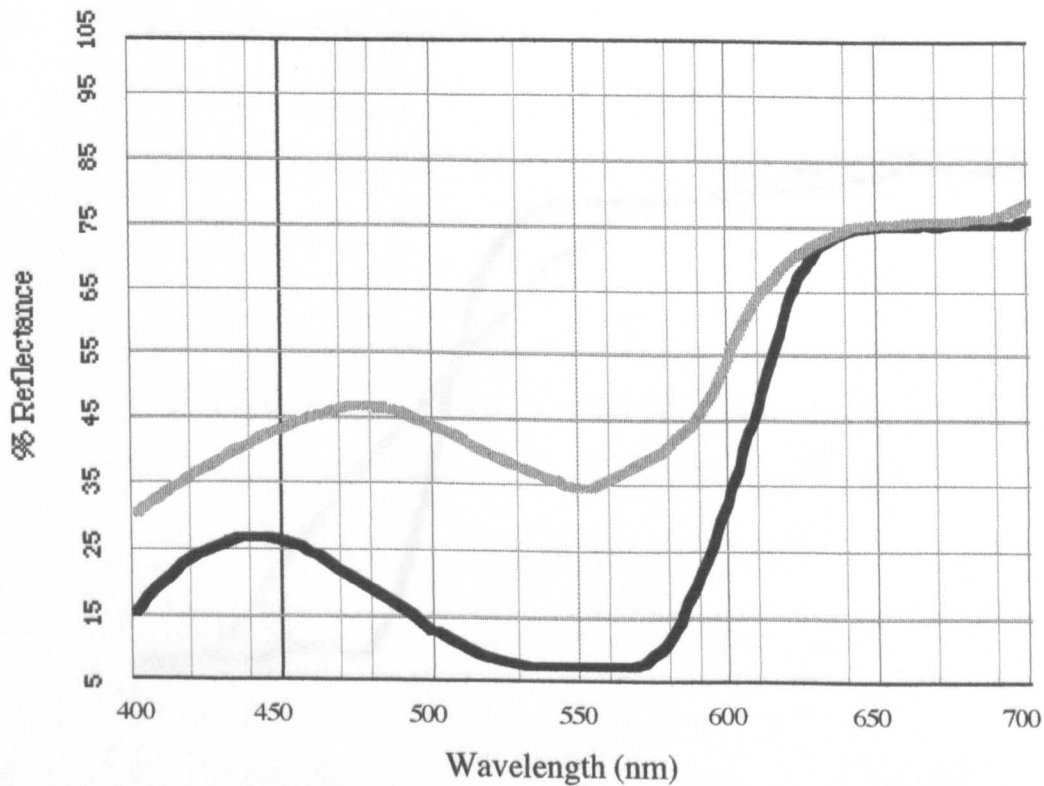


Fig. 2. Spectral reflectance curve of standard process magenta shown in bold, compared with the reflectance curve of an elderberry/mahonia ink sample indicated by the shaded line. Whilst the two samples show similar reflectance at the red end of the spectrum (650 - 700 nm.), they differ considerably in the violet, blue, green and yellow (400 nm - 600 nm.). There is however a general similarity in the overall peaks and troughs, demonstrating that this particular combination may be capable of adjustment.

Tests courtesy of Davison Chemographics Ltd.

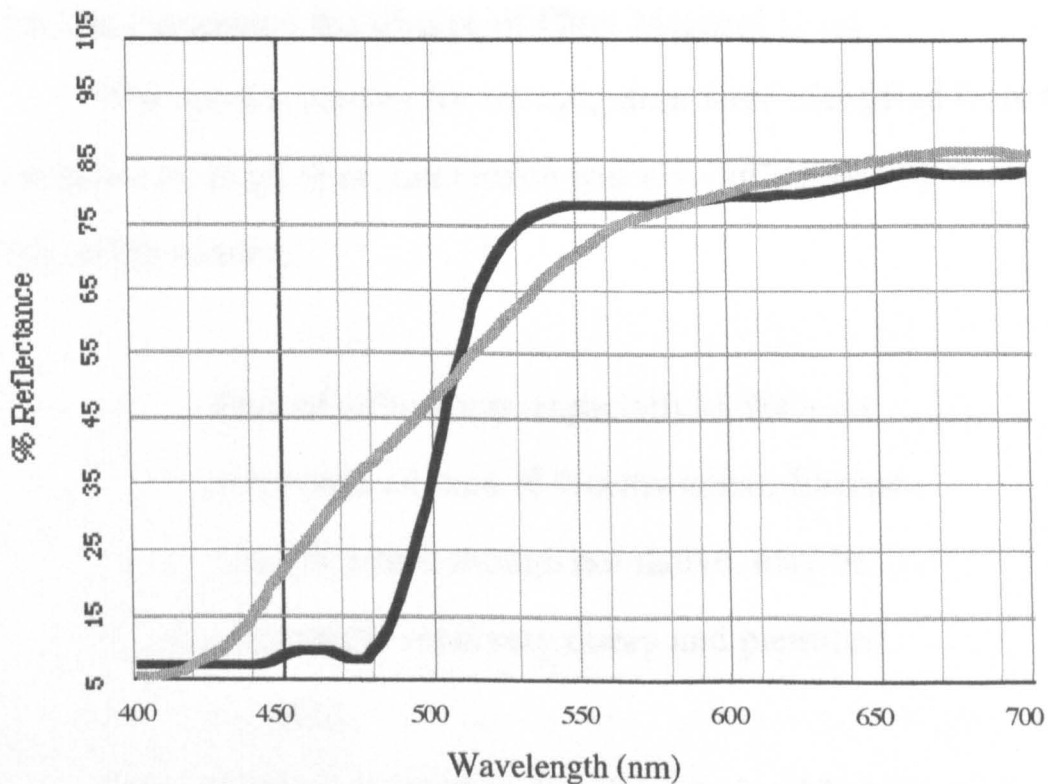


Fig. 3 Spectral reflectance curve of standard process yellow shown in bold, compared with the reflectance curve of a persian berry (buckthorn) ink sample, indicated by the shaded line. As with Fig.2 the vegetable ink sample conforms in general terms to the standard sample, though there is too much reflectance in the blue (430 - 500 nm ) and not enough reflectance in the green (510 - 550 nm). Once again this suggests that adjustments could be made that would bring the vegetable ink sample more in line with the standard. More recent tests with weld have demonstrated, at least to the naked eye, a greater degree of conformity with standard process yellow, though spectrographic tests have not yet been applied.

Tests courtesy of Davison Chemographics Ltd

### Factors Governing the Choice of Plant Material Used

The species chosen for investigation were identified from the literature on plant dyes, but choice was also influenced by the following criteria:

1. Ease of cultivation, especially in the cool temperate climate of Northwestern Europe.
2. Species which though not native, may be obtained in relatively cheap and plentiful supplies.
3. The concentration of colour derived from a given specimen.
4. Their apparent closeness in hue to the process colours
5. Their brilliance, and light fastness.

These factors are not all present in each of the chosen plant sources, Weld for instance is said to be the most permanent of natural yellows, though large quantities of plant material are necessary to obtain sufficient colour for practical use. buckthorn is rich in colour though it is said to be more unstable in light. It is proposed (as a



result of my preliminary tests along with reference to the available literature) that if not one, then some combination of sources may provide an ideal process colour.

By far the greatest body of reference for the work proposed in this study is that contained in the textile dyeing literature, particularly of the 18th and 19th centuries. Reference too, on a more limited scale, is found in the modern work done on food colourings. Reference to vegetable dyes used in the production of inks for printing on paper is less abundant and chiefly consists of passing references to the production of coloured 'lakes' used in 19th century wallpaper manufacture, Japanese wood block printing and some specialised food applications. Along with the necessary botanical and scientific literature, these sources form the main body of reference for the following literature review.

## CHAPTER II

### Yellows

Weld (*Reseda luteola* L. Resedaceae)

The Oxford Dictionary of Botany places weld is a member of a family of some 75 species of annual and biennial herbaceous plants centred around the Mediterranean area but extending to India, Africa and western USA. It grows to a height of 3 to 4 feet. Other members of the family include mignonette (*Reseda odorata*) and the common dock.

It is considered the oldest and most permanent source of yellow dye used in Europe (Rawson, Gardener and Laycock, 1924), the seeds of which have been found in the Neolithic lake villages of modern day Switzerland, where it is presumed they were used for dyeing (Cannon, 1994). Pliny refers to a plant which he calls 'Lutum' which though sometimes translated as 'saffron' more probably refers in this case to the weld used by the Gauls and others dwelling north of the Alps during the time of Julius Caesar (Perkin and Everest, 1918). Robinson (1969) refers to the plants use 'in classical times' when it was known as 'ochra.' Wickens (in Buchanan, 1990) tells us of its exclusive use in the dyeing of Roman wedding garments, and the robes of vestal virgins. Much later it was used as a colourant for the yellow caps worn by European Jews during the middle ages (Cannon, 1994). In

mediaeval Venice the *Capitolaribus de Tinctorium*, the book of dyeing regulations published in 1243, forbids the use of other, less permanent yellows (Cardon, 1990).

Hofenk-De Graaf (1968) claims that its introduction into England was not until the middle ages when it replaced, or was used in combination with Dyers Broom (*Genista tinctoria*) (Cannon, 1994). Archeology has indeed revealed the presence of weld seed in mediaeval sites known to be associated with dyeing. Of the five principal colours of the Bayeux Tapestry, embroidered in southern England sometime before 1082, three - blue-green, olive green and yellow itself, were produced using weld (Duke and Edlin-White, 1993). Interestingly, when the tapestry was examined by conservators in 1983, the colours of the threads on the reverse side were virtually identical to the corresponding facing threads, revealing a remarkable resistance to the effect of light over a period of 900 years (Wilson, 1985). It is possible that considering its widespread occurrence throughout Europe, the use of weld as a colourant in England, as on the continent, could pre-date by some considerable time the mediaeval period.

Bancroft (1813) describes a book translated into English from the Dutch in 1605 entitled, '*A Profitable Booke Declaring Divers*

*Approved Remedies to Take Out Spots and Stains, in Silkes, Velvets, Linnen, and Woollen Clothes; With Divers Colours How to Die Velvets, and Silkes, Linnen, and Woollen, Fustian, and Thread: Also to Dress Leather and to Colour Felles.*' This 'thin and small 4to. volume' (despite its lengthy title) contains, according to Bancroft, the first dye recipes published in England and includes weld as one of only two yellow colourants.

The establishment of Dyers Guilds, and of licences granted to towns like Nottingham and Lincoln (noted for producing the finest green in England, using a mixture of weld, dyers broom (*Genista tinctoria*) and woad (*Isatis tinctoria*) for the production of specific dyes would suggest the existence of written recipes from a much earlier period. The first book on dyeing, according to Bancroft, was the 'Meriegola dell' Arte dei Tentori" of 1429, followed by an improved volume, which also includes reference to the use of weld, dated 1448 and entitled 'Plictho De Larte' also of Venice.

Whatever the true date of its arrival in England, once here it held sway over all other yellows (with the exception of Fustic [*Chlorophora tinctoria*] which gives a somewhat duller shade) until the introduction of Quercitron Oak bark from North America by Bancroft in 1775 (British Patent no.1496 Dated 21.9.1785). Despite

Bancroft in 1775 (British Patent no.1496 Dated 21.9.1785). Despite the superiority of this new dye many English dyers continued to use weld, as Partridge's, 'A Practical Treatise on Dyeing' of 1823 attests:

'Doctor Bancroft has taken much pains to prove that the quercitron, or black oak bark, will give a colour equally as good as the weld; but , English woollen dyers are convinced, after having tried the two, that the weld gives a more beautiful, and more permanent colour...'

In fact Bancroft claimed, in his patent specification, that his new source of yellow produced, '...10 times as much colour as an equal weight of the weld plant...' Whilst it is true that Quercitron does provide considerably more colour than weld, weld has a cleaner, brighter effect, and perhaps more importantly, its fastness to light is the best of all the bright yellow plant colourants (Hummel, 1893-8, Padfield and Landi, 1966). A further point to note is that some writers (Rhind, 1872, Perkin and Everest, 1918 *et al* ) have suggested that the principal source of colour within the weld plant is to be found in its extremities and seeds, and this would suggest that much of the very bulky plant material (the stems and

roots etc) traditionally used is superfluous - the quercitron/weld weight to yield comparisons of Bancroft may therefore be somewhat exaggerated.

In commerce the colour was often sold in the form of an aqueous or powdered extract (Ponting, 1980), and Rhind (1872) describes very thoroughly the process by which 'weld yellow' was produced for use in the paper-hanging trade. He describes a process, which he attributes to Messrs. Collard and Fraser, and which involves the following stages of production:

1. Equal quantities of carbonate of lime (calcium carbonate) and soft water are brought to the boil in a copper vessel and mixed to a uniform consistency.
2. Alum (Potassium aluminium sulphate) added to the solution to the proportion of about 20% and set aside.
3. The weld is placed, with 'roots uppermost' in another copper vessel, in just enough water to cover it, and boiled for 15 minutes, after which the product is filtered.
4. The above is added to the basic solution of water, chalk and alum and brought once more, to the boil. As

soon as the solution has reached boiling point it is removed from the heat and poured into an earthen container in which the colour precipitates in the form of a powder.

5. The supernatant liquid is drawn off and the residuum is placed on large blocks of chalk which in a few hours absorbs the moisture, leaving the colour dry and fit for use.

Although Rhind does not describe it as such, the above is really a description of 'lake' production, that is, a coloured pigment produced by precipitation (of a liquid dyestuff) onto an inorganic substrate such as alumina hydrate, blanc fixe (barium sulphate) or alum (Sanders, 1989). In fact the Collard/Fraser process is very similar to that quoted by Thompson (Trans. Cennini's 'Il Libro Dell' Arte' 1933) of Merrifields translation of the 15th Century 'Bolognese Manuscript' being different only in the addition of white lead, and the use of a hot brick in place of a chalk block for drying. The colour produced here, called 'arzica' was used chiefly by Florentine illuminators.

Lakes were very important in the production of block printing inks in so far as they allowed the normally transparent dye



liquids to become pigmental in form, thus increasing both the strength (though not necessarily the brilliance) and opacity of the resultant ink. Sanders also explains that aluminium hydroxide, 'has a porous open structure capable of absorbing large amounts of dye...when filtered off and dried the resulting pigment has a soft, fluffy texture, unlike the coarse gritty product obtained by drying the dye itself...' The preparation of lakes will be discussed further when we come to consider ink production.

Weld survived, as a commercial product, both the introduction of quercitron yellow (c.1800), and the mid 19th century introduction of synthetics, its last significant use being for the colouring of woollen cloths, braids (for the British army) and silks in the first quarter of the present century (Ponting, 1980). Its use today is largely restricted to craft dyeing though it is reported to have been used in parts of Europe to dye leather as recently as the 1970's (SDC Colour Index 1970). There are no references to its use in printing inks after around 1870.

### Chemistry

The chief pigment, which seems to be specific to weld is the flavone derivative Luteolin (C.I. Natural Yellow 2, C.I. 75590,75580)

being first isolated by Moldenhauer, and subsequently examined by Schutzenberger, Paraf, Hlasiwetz, Perkin *et al* throughout the 19th century (Perkin and Everest, 1918). Luteolin itself is described as being 'hardly coloured' but, in combination with aluminium gives an 'attractive yellow colour' (Swain in Goodwin, 1976). It also contains to a smaller degree the flavone *apigenin* (Cannon, 1994) which also occurs in the seeds and stems of parsley (Furness in Armstrong (Ed.) 1924) and which can be made to produce deeper shades with the addition of chrome and iron salts. Luteolin was finally synthesised by Konstanecki around the turn of the century (Bulletin of the Society of Chemists, Mulhouse, 1901),

### Cultivation

Weld prefers the calcareous soils often associated with derelict sites, slag heaps, road sides and waste ground. Indeed one author claims that too rich a soil only succeeds in producing a tall stalky plant with a reduced colour yield (Partridge, 1823). Rhind (1872) regarded weld as indigenous to Britain, though Cannon (1994) claims its origin to have been in south-central and western Europe. At any rate its cultivation in Britain as a specialist dye plant was well established by the 19th century, plantations being found in

Kent, Herefordshire and about Doncaster in Yorkshire (Rhind, 1872)). Commercially available supplies of dried weld are currently imported from Germany (Dean,1996).

### Nomenclature

Weld is variously referred to as Dyers Weed, Dyers Rocket and Dyers or Wild Mignonette. In France, Gaude; Germany, Gelbkraut; Italy, Erba guada; and in Holland, Woude.

### Buckthorn (*Rhamnus sp.*Rhamnaceae)

The Oxford Dictionary of Botany describes the family Rhamnaceae as 'a cosmopolitan family of trees and shrubs,' they range in size from bushy shrubs to small trees and may be deciduous or evergreen. Of the 875 species of *Rhamnus*, sources name the following as having been used in dyeing; *R.amygdalinus*, *R.oleoides*, *R.saxatilis* (also subspecies *R.tinctorius* ) *R. infectorius*,*R.alaternus*, *R.dahurica*, *R.catharticus*, *Frangula alnus*, *R. Frangula* (Bancroft,1813; Rhind, 1872; Perkin and Everest,1918; Adrosko, 1971; Stuart,1979; Goodwin, 1982 *et al*) though some are not now

considered as necessarily separate species (Cannon, 1994).

Though the bark and ripe fruit of *R.catharticus*, *Frangula alnus*, *R.dahurica* and *R.catharticus* is said to produce colour, it is the fruit, in its unripe state that has been used most extensively in dyeing. In calico printing the so-called Dutch Yellow or Persian Berry Carmine, both lakes produced by precipitation onto aluminium sulphate or calcium carbonate, was used as a pigment. Similarly, Persian berry extract, a concentrated form of the aqueous dye produced by evaporation, was used in the preparation of printing inks during the 19th and early 20th century (Rawson, Gardner and Laycock, 1924).

The dyestuffs known as Persian, French (Avignon) and Turkey berries all consisted of the unripe berries of various species of buckthorn, the names reflecting their place of origin (Rhind, 1872). He goes on to say that the Persian and Turkey varieties were much larger and more sort after by the trade than the French. Certainly the species *R. saxatilis*, found in Persia, is a larger berry, about the size of a pea, and has a 4, rather than 2 celled ovary which forms the fruit. Hofenk De-Graaf (1969) regards this species, along with *R.amygdalinus* and *R. oleoides* as the true source of Persian berries,

while she maintains that *R. alaternus*, *R. infectorius* and *R. catharticus* (a native of Britain) constitute the so-called French berries, or Grain d'Avignon.

Felts produced in Turkestan since prehistoric times probably made use of buckthorn berries as yellow colourants (Berezneva, 1976) indeed fragments of felt produced in Catal Huyuk, in neighbouring Turkey, have been given radio-carbon dates of around 5867 (Mellaart in Burkett, 1979). Hoffenk De-Graaf (1969) describes the use of *R. infectorius* to produce purple on wool in combination with henbane and Lupin in 1st century Rome (from the Papyrus Holmiensis) and this suggests the use of ripe fruit, the juice of which is purple.

Buckthorn berries were much used by dyers from the mediaeval period on. Cannon (1994) tells us of their use in the dyeing of the yellow caps worn by Jews of the period (these caps seem to have been dyed with almost any available yellow dye). Thompson (1933) is of the opinion that Cennini's 'prugnole salvatiche' found in his 'Ill Libro Dell' Arte' (15th century) refers to a species of Rhamnus and not Prunus (as the translation suggests) since it is said to produce a yellow/green artists pigment in combination with weld - a colour unlikely to be obtained by any known species of plum. Sap Green, an artists pigment, now produced synthetically, was prepared from the

ripe berries of *R. catharticus*. Gerards herbal, of 1597 describes the process thus:

'There is pressed forth of the ripe berries a juice which being boyled with a little alum is used of painters for a deepe greene which they do call Sap greene.'

An early 20th century description (Rawson, Gardner and Laycock, 1924) of sap green omits the use of alum, but says that the purple juice was combined with alkali to produce a bright, leafy green. Its use continued as a food colourant, a stain for paper and in calico printing well into the 20th century Stephenson (1989) maintains that sap green is 'of dubious quality' which probably means that it is fugitive to light, though the yellows obtained from the unripe buckthorn berries, particularly when combined with potassium dichromate, appear to have excellent resistance to the action of light (Hummell, 1893-8).

Rhind (1872) relates the experience of a traveller on an ocean passage from Smyrna who noticed that when the pumps were applied after the ship sprang a leak, the deck soon became awash with a bright yellow dye from the bags of Persian berries being stored in the hold.

He concludes from this that the action of salt water potentiates the release of colour, though I can find no other reference to this effect.

Cardon (1990), describes a rather interesting fact regarding the preparation of colour from buckthorn berries. The control of enzymes present in the berries, particularly *rhamnase*, is, she says, crucial to the eventual hue obtained. If an aqueous solution containing the berries is brought slowly to a temperature of 37° C, held there for upwards of 60 minutes, then allowed to come to the boil slowly, the yellow obtained will be a deeper, almost brassy yellow, when combined with stannous chloride (tin). If, on the other hand, the berries are simply plunged into boiling water and simmered for a similar period, the yellow obtained will be a strong lemon, when combined with the same salt. The different results are caused by the action of the aforementioned enzymes which are destroyed by boiling water, while 37°C is the optimum temperature for their influential effect (Cannon, 1994). This point is interesting in so far as the preparation of process printing inks are concerned. Because the standard commercial process yellow is a slightly greenish lemon, the latter of Cardons methods would seem to be imperative for satisfactory results.

As has already been stated, buckthorn yellows, particularly

those obtained from *R. Saxatilis* perform relatively well in light. Besides the 'very fast' quality described by Hummell, tests performed on buckthorn in combination with a variety of metallic salts show a range of permanencies (Padfield and Landi, 1966). The addition of copper, in the form of copper sulphate, is reckoned, along with potassium dichromate, to give greatest fastness (LF 4-5 BS 1006). Aluminium sulphate, is said by Dalby (1985) to equal copper and chrome, and by others to be inferior to them (Padfield and Landi, 1966). The result of the introduction of stannous chloride (tin) to the dyestuff, is described as 'only fairly fast to light' by Perkin and Everest (1918). These assessments all refer to the action of light on dyed textiles, tests on printed papers and fabrics may well differ, depending on the ink deposit constitution and testing of printed samples will form part of this study. Lakes for instance will almost certainly give different degrees of light fastness than simple aqueous extracts thickened with starches and gums.

### Chemistry

Though the colour derived from the various unripe buckthorn berries may be broadly similar the chemical pigments present can vary. Of the species providing Persian berries, Perkin and Everest



(1918) identified three members of the flavonol group, namely, rhamnetin (C.I. 75690), rhamnazin (C.I. 75700) and quercetin (C.I. 75670). Cannon (1994) adds the anthraquinone, emodin (C.I. 75430), and another flavonol, Kaempferol (C.I. 75640) to this list, though Hoffenk De-Graaf (1969) states that this pigment is found only in *R. catharticus*. Whilst it used to be thought that rhamnatin was the principal pigment in Persian berries (Perkin and Everest, 1918, Rawson Gardner and Laycock, 1924 *et al*) the Colour Index (JSDC, 1979) maintains that kaempferol is the chief constituent, and Cannon (1994) certainly includes it as one of the 4 main pigments. In addition the index indicates the presence of xanthorhamnin (C.I. 75695), kaempferol 7-methyl ether (C.I. 75650) and several other minor pigments. The various buckthorn yellows are listed in the index as Natural Yellow 13, 14 and 15.

It is not surprising, with so many species providing similar colours along with the difficulty of identifying sources which may contain a mixture of species, that researchers have held differing opinions regarding their chemical make up.

Interestingly, the species *R. catharticus* is said by Stuart (1979) to contain a fluorescent pigment, rhamnoflourin, but neglects to say which part of the plant contains it, though it quite possibly exists in

both bark and fruit. Rhamnatin can also be found in *Delphinium zalil* which is indigenous to Afghanistan, after gathering, it is sent to India, where it is used in silk dyeing ( Hofenk De-Graaf, 1969).

### Cultivation

*Rhamnus catharticus* appears to be native to Britain, according to the Field Guide to Trees and Shrubs of Great Britain (1981) it grows wild in chalk and limestone soils in central and southern England. Stuart (1979) notes its presence in hedgerows, and goes on to say that cuttings taken in summer are easily rooted. Specimens can be bought along with *R. alaternus* and *R. frangula* from specialist nurseries (Philip, 1995). Cannon (1994) does not recommend cultivation of the latter in cold inland areas, but says that it thrives well in maritime or urban locations. The unripe berries, he goes on, should be picked when fully grown, but before they have ripened to become black in July or August. The species *R. saxatilis*, *R.amygdalinus* and *R. oleoides* the true persian berries are probably native to Persia and parts of Asia Minor (Anatolia) (Rhind, 1872) though a specimen of *R. tinctorius*, said to be sub- species of *R.saxatilis* can be seen growing unprotected at Kew. The species *R.infectorius*, which along with *R. alaternus*, and *R. catharticus*

*R. infectorius*, which along with *R. alaternus*, and *R. catharticus* constitute the French berries, grow wild in Spain and southern France, though they appear to be quite at home in southern England. Bean (1980) includes *Rhamnus infectorius* and *R. tinctorius* as being both hardy in the British Isles. Commercial sources currently available are chiefly imported from India (Dean, 1996).

### Nomenclature

Berries from the Buckthorn family have been referred to in Britain as, Persian Berries, Dyers Buckthorn, Yellow Berries, Hungarian Berries. In France, Grain d'Avignon; Germany, Gelbeere; Italy, Spincervino and Grani d'Avignon; Holland, Persische bessen.

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## CHAPTER III

### Reds

Madder *Rubia tinctoria* L. Rubiaceae

Madder is a member of one of the largest families of flowering plants, being one of over 10,000 species (Oxford Dictionary of botany, 1994), about 35 species of madder have colour producing capacity according to Wickens (1983). Among those which are capable of providing reds suitable for dyeing and printing, are *Rubia cordifolia* and *Rubia munjista*, both native to India, including the Himalayas, and much of Asia, as well as tropical Africa (Cannon, 1994). In the case of *Rubia cordifolia* the whole plant is lifted and used complete to obtain colour (Hecht, 1989). *Rubia peregrina*, the wild madder found in southern parts of Britain, provides, according to some authors, an inferior pink dye (Ponting, 1980, Stuart, 1979, Duke & Edlin-White *et al*), and by no means as prolific as its asian relatives (Cannon, 1994). Others however, state that it gives a brighter colour than *Rubia tinctoria* (Rawson, Gardner & Laycock, 1926), though this may simply refer to the fact that pink could be regarded as a 'brighter' colour than red, whilst at the same time being infinitely less intense.

Lady's bedstraw, Hedge bedstraw and Northern bedstraw (*Galium verum*, *Galium mollugo* and *Galium boreale*), Sweet woodruff (*Galium odoratum*) Dyers woodruff (*Asperula tinctoria*)



and Goosegrass or cleavers (*Galium aparine*) are all found throughout Europe and provide similar shades to madder proper (Rhind, 1872; Rawson Gardner and Laycock, 1924; Goodwin, 1982 *et al*).

*Rubia tinctoria*, is a climbing perennial, growing to a height of about three feet and can sometimes be found naturalised in Britain in hedgerows and thickets, or scrambling through vegetation (Cannon, 1994). Numerous small yellow-green flowers each with four petals appear from mid-summer onwards, followed by globus purple-black berries in the autumn.

It is the root of this, and related plants that has provided a red dye throughout history, probably since the time of the great civilisation in the Indus Valley some 3,000 years B.C.(Adrosco,1972). Subsequent workers were able to derive a range of colours from madder including, black, violet and puce, by the intervention of chemical modifiers, thus extending the versatility of the plant, (Crookes, 1874).

Other ancient finds include linen dyed with madder in the Egyptian tombs of the Nile valley, where Herodotus describes its use there around 450 BC (Ponting, 1980). Pliny also mentions its use by the Persians as well as the Hindus and Egyptians in the first century

AD (Historia Naturalis Book XIX, ch. 17) ). The oldest written recipe for a madder dye appears in the 3rd century 'Papyrus Graecus Holmiensus'. Virgyl, according to Rawson, Gardner and Laycock (1926), describes the tinctorial effect upon pigs bones after having being fed with madder. The same authors maintain that madder was probably first grown in Europe in Spain, having been introduced from Algeria at the time of the crusades, though Pliny's reference to madder cultivation in the suburbs of Rome places its European debut much earlier, adding that '..nearly all our provinces ... produce it in great abundance..' It may be that the plant was, in fact, reintroduced into Europe during the middle ages after a period of disuse, its earliest recorded post Roman Empire cultivation being at St. Denis, near Paris, in the 7th century (Hofenck De-Graaf,1969). Madder was grown in Zealand (Holland) as early as the 10th century (Rawson, Gardner and Laycock, 1924) and it was this source which was eventually to become prized above all other European products by the time of the 18th century. Berthollet (1823) regarded the madder grown in the lower Rhine as being equal to the Zealand product, and this is of some interest when considering conditions for ideal cultivation.

Partridge (1823), an English dyer working in America,

describes four qualities of mainly Dutch imported madder, distinguished in the commerce of the time; 'crop', 'umbro', 'gamene' and 'mull'. The Dutch themselves seem to have distinguished only three categories, Krap, Geneen and Mull (Duke and Edlin White, 1993). Of these 'Krap' 'crop' or 'Kor Kraps' was considered best, being the very central part of the root. The outer parts (mull) being of poorest quality, used chiefly for the production of dull reds and 'dirty drabs'. The roots were separated into these qualities by a series of poundings and riddlings after being dried in ovens (Adrosko, 1971).

The finest madder was said by most 18th and 19th century dyers to come from the eastern Mediterranean, where it was imported into Europe via the ports of Trieste and Leghorn, though Ponting (1980) suggests that this belief was probably based on the fallacious assumption that the superior reds found on cloths dyed in that region were the result of a superior dye source, and not superior craftsmanship. He maintains that the plant itself, being identical, was perhaps processed with greater skill in the east than in Europe, and this, in fact is quite evident from the examples of dyeware we have from that area (see *Origins of Chintz*, Irwin and Brett, 1970 for superb examples of Indian Madder reds). Certainly, the red achieved

in India during the 17th and 18th centuries, were far more intense and permanent than anything produced in the west, involving a prolonged and complicated method of manufacture, though this may also be due to the fact that the Indian variety was largely produced from *Rubia cordifolia* (Irwin and Brett, 1970). The Indian technique was probably the real reason for the superiority of colour derived from *Rubia tinctoria* in the eastern Mediterranean. The complex preparation of the colourant known as Levant or Adrianapole red and later styled as Turkey Red, was derived by the Turks from the Indian technique and involved some 16 separate operations over a period of a month, or more (Ponting, 1980). Hoffenk De-Graaf (1969) lists 22 separate stages to its production. The colours achieved using this process were among the brightest and most permanent dyes available.

In 1790 the technique was eventually brought to Scotland via Rouen in France which had in turn been transported there by Greek workers who were familiar with the Turkish process (Rawson, Gardner and Laycock, 1924). The details of the preparation of Turkey red are not within the scope of this study, since they apply more specifically to fabric dyeing and resist printing, but it is worth noting that the colour swatches appearing in Crookes, 'A Practical Handbook of Dyeing and Printing' published in 1874, reveal just how

superior this technique was, even compared with the synthetic alizarin (one of the principal pigments present in madder) then being used. The sample printed in Turkey red is, by any standard, much brighter and cleaner than the synthetic version which appears below it, what is more, the synthetic swatch has not survived the 122 years since they were placed in the book without some deterioration (patchiness and some discolouration), none of which is evident in the madder. Indeed the use of this technique survived well into the 20th century, though by then synthetic alizarin was being used in place of madder (Ponting, 1980).

The production of a red ink from madder is not without its problems, mainly due to the variety of pigments present in the root. Besides red and a smaller amount of a purple colourant, madder also contains quite large quantities of yellow and brown (see 'Chemistry' below) and these can very easily dominate the final extract, turning what should be a good scarlet or pink to a dull brick red (Crookes, 1974). A variety of techniques have been developed over the years to combat this tendency, most of which involve careful control of temperature, and the addition of calcareous matter. Authors favouring a maximum temperature of 60<sup>o</sup> C and the addition of chalk to the dyebath include, Duke and Edlin-White (1993); Goodwin

(1980); Adrosko (1972) and Lesch (1970), though the latter suggests that the extract may be obtained at temperatures almost approaching boiling point, while Dalby (1985) and Beat (1994) do not mention the use of lime but agree on temperature. Cannon (1994) agrees with the addition of slaked lime but suggests a maximum temperature of 85° C.

Wickens (1983) suggests a method of extraction differing in a number of points from those authors already mentioned. Her method, requires the addition of madder to a pre-heated quantity of hard water (30° C), the temperature then raised slowly (over a period of 90 mins) to boiling, and at that point reduced immediately to 90° C.

Besides these aqueous extraction methods a number of other procedures have been employed to yield higher proportions of colour from madder roots and to eliminate the various useless colourants present. Crookes (1874) describes a variety of processes mainly developed in France and these include the fermentation process used to obtain a colourant known as fleur de garance, or flowers of madder (Julian and Rouer, 1851), Pernods hydrochloric or sulphuric acid process, involving the use of one to two kilos of concentrated acid to every 100 kilos of madder, the purpose of which was to

saturate the calcareous matter present in the root. Garancin, originally known as flowers of lagier, was a product related to the above, developed and patented by Lagier, Colin and Robiquet in 1828 and involved much higher proportions of acid, the purpose of which was to destroy not only the calcareous matter, but to rid a proportion of the fibrous woody material as well as the useless colourants contained in the root. The increase in colour derived by this method was described by Crookes as 'a real gain.' Other products included Garanceux, another acid process, but applied to what was called 'spent madder' ie. madder which had already been through an extractive process, but which still contained useful colourant.

The introduction of acids, except in quantities far smaller than those described above are not processes which one would wish to reintroduce as part of an environmentally friendly means of manufacture. Likewise, the procedure described by Rawson, Gardner and Laycock (1926) for obtaining a madder lake which included the use of lead acetate, would seem to be undesirable for the same reasons. Alternatives, must therefore be sought and there are procedures outlined in the literature which might profitably be pursued.

For the production of madder lakes suitable for printing inks

which do not involve acidic processes or toxic metal salts, we may turn to the apparently simple 19th century procedure developed by Englefield and described by Crookes (1872). This consisted of a series of five compressions of 2 oz of root contained in a muslin bag in successive quantities of cold water. The liquid is poured off after each pressing and the total extract is heated in a silver or stoneware vessel to near boiling point. A solution of aluminium sulphate (1oz to 1 pint of water) is added followed by 1.5 oz of saturated sodium carbonate solution. The product is washed, first by decantation and then filtration to produce 1 oz of pigment. This procedure approximates to that described by Stinchechun (1983/4) as being the method by which the Japanese extract colour from madder. Interestingly, the Japanese are said by the same author to remove the yellow pigment in *Rubia cordifolia* by means of a pre-soak in rice.

As has already been mentioned, madder reds are among the most permanent colours derived from plant material. Padfield and Landi (1966) give approximate BS ratings of between 5 and 7 after 25 million lux hours of natural light for wool dyed with madder mordanted with aluminium sulphate, with similarly impressive figures for artificial light. Dalby (1985) gives BS ratings of between 4 and 5 for madder combined with aluminium sulphate and cream of



tartar, and stannous chloride and oxalic acid. Stephenson (1989) gives a BS rating of 6 for the various madder lakes prepared with alumina bases. The literature on lightfastness is perhaps misleading in so far as comparison of results are concerned, certainly from the point of view of this study. Most lightfastness ratings are concerned with the effect of light on dyed textiles, and as such may not be entirely relied upon with regard to printed paper applications. The exposure times vary greatly from one study to another, and most authors, with the exception of Stephenson, give what they describe as approximate BS equivalents.

### Chemistry

The principal pigments found in madder root, according to Cannon (1994), are the anthraquinones alizarin, rubiadin, purpurin, pseudo-purpurin, and mungistin. The isolation of the most important pigments, alizarin and purpurin, was first achieved by Robiquet and Colin in 1826 and 1828 (Perkin and Everest, 1918). Alizarin was subsequently synthesised from anthracene (coal tar), first by Graebe and Liebermann in 1868, followed by Perkins improved method in 1869 (Fox, 1987). This improved procedure became the basis for subsequent commercial production (Rawson, Gardner and Laycock,

1926). Of the two main colouring principals derived from madder, Alizarin is of most importance as a source of red. It exists in the root of *Rubia tinctoria* in the form of the glucoside rubierythric acid (Hofenk De-Graaf, 1969) and together with purpurin forms 1.5 to 2% of the total root (Rawson, Gardner and Laycock, 1926). *Rubia peregrina* (wild madder) according to Cannon (1994) contains pigments which include purpurin but not alizarin. In practice, the two pigments exist in most madder preparations and the Colour Index lists madder as Natural Red 8, along with a range of madder lakes, obtained by the use of acidic and fermentitive processes (Garancine, Garanceux etc), as Natural Red 9, 10, 11, and 12 (CI 75330, 75420).

### Cultivation

Both *Rubia tinctoria* and *Rubia peregrina* (wild madder) can be cultivated quite easily in Britain, particularly in southern and central regions, although cultivation on a commercial scale has never proved economic (Rhind, 1872). The ideal soil conditions for cultivation are said by some authors (including the most recent references) to be as follows; well drained, well cultivated, calcareous soil (Cannon, 1994, Dalby, 1982, Ponting, 1980, Stuart, 1979). However, the high quality commercial madder grown in Holland

right up until the introduction of synthetic alizarin (and beyond) might suggest otherwise. Whilst the soil should ideally be rather friable, to aid harvesting of the root, 19th century analysis of the Zealand soil showed considerably less calcareous matter than that found in the soils of Alsace and Vaucluse, two other important madder growing regions (Crookes, 1872). Rhind (1872) describes the ideal soil conditions as being, 'moderately rich, light and somewhat sandy' with no mention of chalk or lime. Berthollet (1823) says that whilst madder can grow in stiff, clayey soils, it thrives best in 'rich, soft, moist, and very slightly sandy soil.' The finest madder, even beyond that grown in Holland, was cultivated in Smyrna (in modern Turkey) where soil conditions were again quite different.

The common denominator as far as soil is concerned seems to be that it should be light and friable with adequate, but perhaps not too rich a nutritional value. It is interesting to note too, that madder was often grown on land that had once been marshy or even totally submerged, as in the case of Zealand and Vaucluse (Crookes, 1872). Climate, too, seems to be quite variable from one traditional growing area to another.

A glance at the regions named above serves to demonstrate the climatic range within which the plant has thrived. Most authors state

*Rubia tinctoria* should be grown for between 14 and 28 months before attempting to lift the root, this, it is said, improves the colour yield, which becomes more concentrated with time (Dalby, Ponting, Perkin and Everest,). Some suggest longer periods, and Cannon (1994) states that in Britain the plant should be allowed to grow for periods in excess of 36 months before harvesting. Rawson, Gardner and Laycock (1926) state that in Eastern Europe, the roots were not removed from the soil until a period of five years had elapsed.

Propagation from seed, of both *Rubia tinctoria* and *Rubia peregrina*, can take place in spring or autumn or by root division (Stuart, 1979), seedlings and small plants can also be obtained from around a dozen nurseries throughout Britain (Philip, 1996). The roots are lifted in autumn, and a portion retained for replanting (Cannon, 1994).

Once harvested, the roots must be thoroughly dried, either by the action of the sun and air, or as was the case in Holland, by artificial heat (Rawson, Gardner and Laycock, 1926). In Britain, the climate does not usually allow for natural drying of the root, this being best achieved in a slow oven (Cannon, 1994).

The bedstraws (see above) can all be cultivated in Britain and require much the same conditions for growth as madder itself (Dalby,

1982). The madder imported into this country for use by craft dyers is mainly obtained from Iran (Dean, 1995).

### Nomenclature

In England madder has been known by that name since the middle ages, though in Saxon England it was referred to as Grias or Medere (Duke and Edlin-White, 1993).

In France it is called Garance; Germany, Farberrote; Italy, Robbia; Holland, Meekrap. The root itself, according to Crookes (1872) was referred in in many countries as lizari' or 'alizari' and Hoffenk De-Graaf (1969) states that these terms are of Persian origin.

### Pokeweed (*Phytolacca americana*) Phytolaccaceae)

According to the Oxford Dictionary of Botany, *Phytolacca* is a genus of trees shrubs and herbs of which there are 25 species, though some authors suggest as many as 35, confined mainly to warm and tropical climates. Cannon (1994), describes it as a native of the USA extending south to Mexico, being grown also in Europe as an ornamental, and as a source of red dye. It grows to a height of between three to ten feet, having purple veined stems and bearing

green or pink star shaped flowers borne on erect and spreading spikes, giving rise to purple/black ten lobed berries. Stuart (1979) describes the plant as having white or sometimes pinkish flowers followed by purple reddish berries, and goes on to describe several interesting medicinal properties which have been the subject of recent investigation, including its ability to destroy snails as a possible means for controlling the tropical disease Bilharzia.

Stuart (1979) says that the plant was introduced to American settlers by Indians who knew it as *pocan* or *cocum* from which its common name was derived. Collins English Dictionary describes this plant as being associated with the Algonquian speaking tribes, who referred to it as 'puccon' though this name seems also to have been used for other sources of red dye such as *Lithospermum incisum* and *L. caroliniense*. The leaves of *Phytolacca dodecandra* known locally as Poko or Ipoko, are used by craft dyers in southern Africa to produce beige, greens and browns (Noy, 1988).

Of the species which form the basis of this investigation, pokeweed is the only one possessing toxic properties. Tampion (1977) states that the root is the most poisonous part of the plant, but adds that, 'the whole plant is best considered potentially dangerous.' Again, Stuart describes it as 'toxic and dangerous,' as does the *Readers Digest*

*Digest Encyclopaedia of Garden Plants and Flowers*. Cannon (1994) states that the root, stem and seeds are toxic, and that children are said to have been poisoned by eating the fruits, though he does not go on to recommend any special precautions when handling the fruit. Goodwin (1982) and Lesch (1970) make no reference to the poisonous nature of the fruit in their dye manuals, and therefore suggest no special precautionary procedures.

The historical use of pokeweed as a dye or pigment is not well documented, though the North American Indians are known to have used the root for medicinal purposes (Stuart, 1979). None of the 18th and 19th century authors, Bancroft, Berthollet, Partridge, Crookes *et al*, make mention of its use, nor is any reference to pokeweed found in the early twentieth century works by Rawson, Gardener and Laycock (1924) or the seminal work on natural organic colours by Perkin and Everest (1918). Ramsey, in his *History of South Carolina* (1809, vol 2, pp249-252) notes:

'..the juice of poke berries boiled in rainwater and set with alum, imparts to wool a beautiful crimson ..'

More recent references, found largely in the 'craft dyeing' literature suggest an interesting, but rather light sensitive colour

derived from this plant (Cannon, Goodwin, Adrosco, Lesch). Sweeney, writing in *The Herberist* (vol 10, 1944), states that the colour kept in a liquid state soon turns brown, but that she avoided this tendency by allowing the liquid to evaporate, leaving a dry powder, which returned to red with the addition of water. She also quotes Porcher as recommending the addition of alum (aluminium sulphate) as a colour fixative, and boiling with sugar and alum as another method by which the colour can be made more permanent (Resources of Southern Fields, 1863). Cannon (1994) maintains that a range of reds, pinks and magentas can be derived from pokeweed by adding acid to an infusion of the berries. In my initial tests the addition of citric acid appeared the most effective way to produce magenta. Stuart (1979) refers to its use as an artists paint , but provides no other details. The same author mention its use as a colourant (after special treatment) for wine and confectionery, though it has no designated 'E' number as a permitted food additive in Europe. The Colour Index makes no reference to pokeweed.

### Chemistry

The chemical make-up of the berries of pokeweed is said by both Cannon (1994) and Stuart (1979) to be not fully understood,



though Stuart describes the following substances as being present; Saponins (to 25 per cent); mucilage; tannic acid; phytolaccinic acid; red pigment; caryophylline. Cannon states that the pigment present is 'thought to be the anthocyanin, cyanidin'. If this is correct, then the colourant can also be found in a number of other fruits, such as cranberry (*Vaccinium vitis*), sloe (*Prunus spinosa*), red current (*Ribes rubrum*) and raspberry (Perkin and Everest, 1918). Cardon (1990) describes the presence of Phytolaccanoside, a pigment identical to that found in beetroot and thought to represent 95% of the colouring power of the berries. Cardon goes on to describe, however, the composition of pigments found in Pokeweed berries as reported by Russian analysts (Chumbalov and Mukhamedyarov, 1969) as being somewhat different. These workers report 'the presence of an anthocyanin heteroside of cyanidol (9.26% of dry weight) which would seem something of a contradiction. My own initial extractions do show a considerably greater concentration of pigment in pokeweed berries than either the anthocyanin containing fruits mentioned above or the rather weak concentrations of colour found in beetroot. In addition it should be noted that of the above mentioned species, only pokeweed seems to have the capacity to produce a true magenta hue (see colour test swatches).

### Cultivation

Pokeweed is grown on a market garden scale in Carolina and in other parts of the USA, producing a winter crop of edible vegetable leaf Stuart (1979). It prefers a rich, light, well drained soil in a sunny position (Cannon, 1994). Small plants, available from around a dozen nurseries throughout Britain (Philip, 1996), may be planted out in March to produce berries suitable for ink production in late August and September, the stems should then be cut down in November.

### Nomenclature

Pokeweed is also known as pokeberry, pokeroot and the 'red ink plant,' but no other names appear to have been used to describe it, apart from those Indian terms referred to above.

### Elder (*Sambucus nigra* L. Caprifoliaceae)

There are, according to various authors, between 20 and 40 species of elder, of which *Sambucus nigra* and *Sambucus ebulus* (dwarf elder) are said to produce usable pigments. The Oxford Dictionary of Botany describes the genus as consisting of deciduous trees, shrubs and herbs with pithy stems with panicle-like clusters of

flowers developing into berries or 'drupes,' which in the case of *S. nigra* are edible. The tree grows to a height of up to 30 feet and is regarded by most authors as native to Britain and Europe, North Africa and western Asia. Rhind (1872) describes the growth of elder as being with 'singular rapidity.' More recently Cannon (1994) tells of its distinct preference for alkaline and nitrogen-rich soils such as can be found around badger sets and rabbit warrens, and it is common to see elder growing around derelict buildings where soil conditions can often be similar.

The pharmacological uses traditionally ascribed to elder are noted by many authors including Vickery (1995) and Stuart (1979), the former describing seven different 'beneficial' effects valued in folk medicine. The same author notes too its use as an insect repellent, being often planted around toilets to prevent infestation by flies.

The berries of *Sambucus nigra* and *S. ebulus* have been used as a source of colour since early times, Stuart (1979) records the use of Dwarf or Danes elder (*S. ebulus*) as a blue stain by the Anglo-Saxons and Gauls, though Cannon (1994) suggests that this belief may be open to question (whilst the berries most certainly do provide a blue dye, Cannon feels that for chemical reasons it is more likely that the plant in question was woad [*Isatis tinctoria*]). *Sambucus nigra*

appears in many dye manuals as a source of mauves, pinks, reds and slate blues (Mairet,1952; Lesch,1970; Adrosko,1970; McGrath, 1977; Goodwin,1982; Wickens,1983; Dalby,1985; Cannon,1994 *et al*).

*The Field guide to the Trees and Shrubs of Great Britain* (Readers Digest,1981) states that a black dye was obtained from the bark, green from the leaves and blue or lilac from the flowers. It makes no mention of the dye contained in the fruit.

As with pokeweed however, the technical literature of the 18th, 19th and early 20th centuries, including Berthollet, Bancroft, Partridge, Crookes, Perkin and Everest, Rawson, Gardner and Laycock, Ponting *et al* , are all silent on the subject. The reason for this absence from the literature is no doubt due to the relatively poor light fastness attributed to most of the *anthocyanin* pigments present in fruits (see also 'Garden Huckleberry'). My own tests have shown that with careful use of acidic modifiers a reasonable process magenta can be derived from elderberries (*Printmaking Today* Vol 4, No 4, pp 26-27) and with better than might be expected light fastness, though controlled tests have not yet been initiated.

It is true that a number of authors describe elderberries as having poor light resistant properties, among them Dalby (1985) who gives BS ratings of only 2 to 3 for very pale pinks obtained in

combination with 1 per cent *Potassium dichromate* or 2 per cent *copper sulphate* along with formic and acetic acids. Cannon (1994) suggests the addition of *copper sulphate* achieves faster colours, which appear (in reproduction) to be nearer to dark brown than magenta, thus rendering this particular additive unsuitable for the purpose of this study. Lesch (1970) however maintains that elderberries, used fresh, and combined with *aluminium sulphate* will produce a range of fast purples. She adds that the berries should ideally be 'very ripe' and this observation along with her results, accords with my own results.

### Chemistry

The pigments contained in the berries, according to Cannon (1994) are the anthocyanins Chrysanthemun and sambucin, which are derivatives of quercetagenin. A more detailed analysis of the pigments present in elderberry derived from the work of Reicheln and Reichwald (1960,1977) and Harborne (1967), accords with the above (Timberlake and Bridle in Markakis,1982).The berries also contain some tannins, thus rendering them capable of producing quite deep blacks when combined with *ferrous sulphate*.

### Cultivation

Elder grows freely in most soil conditions, but prospers best in 'any good garden soil' according to Cannon (1994). Stuart (1979) is more specific in suggesting that elder thrives in moist, nitrogen rich soil. It should be planted between October and March in sun or partial shade, and pruning should be avoided as this reduces inflorescence and subsequent fruiting, which latter takes place in September. Propagation is possible from hardwood cuttings (October/November) or from half ripe shoots with a heel, taken in July or August .

### Nomenclature

Elder has been variously referred to as Boor or Bour tree (Vickery,1995) and by 'the ancients' as rixus, ixus atke and most commonly, sambucus, which gives rise to its modern generic name (Stuart, 1979).

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## CHAPTER IV

### Blues

### Indigo (*Indigofera tinctoria* Leguminosae)

Blue is by far the rarest of all the colours obtainable from vegetable sources. The plant species responsible for most of the blue dyes and pigments used since earliest times are those belonging to the genus *Indigofera*, of which there are according to Cannon (1994) over 700 species, occurring mainly in the tropics. The Oxford Dictionary of Botany states that the species is adapted to dry conditions and is found in all arid zones of the world. Indigo grows to a height of around 3 feet, with red to violet flowers crowded into spikes, the seeds are contained in cylindrical pods, which explode to disperse. Most authors agree that

*I. tinctoria* originates from India, where traditionally, the best quality sources were located. Several authors (Rhind, 1872; Perkin and Everest, 1918; Mairet, 1944; Adrosko, 1971 Goodwin, 1982; ; Van Stralen, 1993; Cannon, 1994, Stinchechum, 1983/4 *et al* ) name other members of the same genus producing chemically identical pigments are *Indigofera sufruticosa*, *I. arrecta*, *I. argentea*, *I. heterantha* (*syn. I. gerardiana*). They describe too, several other unrelated plant species producing the same blue pigment including, *Marsdenia erecta*, *Wrightia tinctoria*, *Strobilanthes flaccidifolius*, *Tephrosia toxicaria*, *Polygonum tinctoria*, *P. chinense*, *P.*

*Lonchocarpus cyanescens*, *Nerium tinctorium* (syn. *N. oleander*), *Mercurialis anna*, *Mercurialis leiocarpa* and the plant famously used as a body paint by ancient Britons, Woad (*Isatis tinctoria*) (Rhind, 1872).

Despite this proliferation of sources, and admitting that some variation of hue is obtained from them, they nevertheless provide substantially the same chemical pigments (Perkin and Everest, 1918). Few other plants produce blues of a distinctly different chemical composition, and hence the relatively limited range of available vegetable blues.

Concentrating on *Indigofera tinctoria*, Perkin and Everest (1918) note the existence of records in Sanskrit describing methods of preparation dating from 'remote antiquity' (about the 4th century BC according to Cannon, 1994), we find reference too, to its use 5000 years ago, where, according to Rawson, Gardner and Laycock (1926) it was used to dye ribbons found on Egyptian mummies. Adrosco (1971) speaks of an ink, known as indicum, produced by the Romans from indigo, and in the first century AD, Pliny (*Historia Naturalis* Books 33 ch 57 and 35, ch 26) refers to two types of Indigo, one of which which he erroneously describes as '..a slime

which adheres to the scum upon reeds.' Though he may be confusing Tyrian Purple (derived from a species of mollusc, and which according to Prescott and Ridge [1933] produces a pigment identical in structure) it is thought that at least one of his references is true indigo, since he states that its origin is Indian, and that it burns with a purple flame, the latter being an accepted test for the determination of the presence of indigo (Crookes, 1872, Sherwood Taylor, 1933).

Most authors (Rhind, 1872; Perkin and Everest, 1918; Adrosko, 1971; Ponting, 1980; Goodwin, 1982 *et al*) claim its introduction into Europe as dating from the early to mid 16th century, though due to opposition from Woad (*Isatis tinctoria*) growers, backed by several royal edicts throughout Europe, banning its use, it did not come into open production until more than a century later (Goodwin, 1982). Despite these attempts to ban indigo dyeing, Crookes (1872) notes the importation of 500,000 pounds of indigo into Amsterdam as early as 1631.

Once indigo became accepted in Europe, first Spain, and then Britain in particular, made great efforts to establish plantations in their New World possessions. With the loss of the North American colonies, indigo cultivation declined, and India, once again became the main producer (Ponting, 1980). Fox (1987) quotes the *British*

*Almanac* for 1863, which records the British consumption of indigo for that year standing at 85,000 cwt (4250 tons). Berthollet (1791) describes in great detail the various sources of, and manufacturing processes connected with indigo, as does Bancroft in his, *Experimental Researches Concerning the Philosophy of Permanent Colours* (1794 and 1813).

The methods by which indigo can be processed into usable blue colourants remained substantially the same for almost a century after the above authors outlined them. The so-called vat method, is essentially as follows: The plant is steeped in water to allow fermentation to take place, the solution then being oxidised with air in a separate vessel. Large vats were used to ferment the plant material (up to 1000 cubic feet capacity) followed by precipitation vats, of smaller dimensions, in which air was introduced to the solution by means of paddles, pumps or the so-called 'shower bath' method. This causes precipitation of the pigment, which after the supernatant liquid is drawn off, is dried and formed into cakes, ready for sale. Methods of extraction, using heated water ( 150 to 160 °F) are also described in Bancroft (after Roxburgh, 1797) and this method required the exclusion of air during the the heating process.

As with madder, the introduction of acids to the process during the early 19th cent was well understood, Berthollet (1791), Bancroft (1794 and 1813) and Partridge (1823) all describe the preparation of a dye substance known as 'chemick' which involved the use of sulphuric acid. Adrosko (1971) quotes Hazericks description of the discovery of chemick by Seidelman in the mid 18th century in Altenburg, Saxony, giving rise to its other name 'Saxon blue.' Later, an improved version of this product, known as sulphonated natural indigo (CI 75781 Natural Blue 3) was developed, producing a brighter blue (Ponting, 1980).

The rather complicated procedure required to colour cloth with indigo arises out of the fact that indican, a colourless and water soluble compound of indoxyl found in the leaves of the plant, becomes the blue insoluble compound indigotin, when exposed to air. This fact makes adherence to fabric or yarn difficult without the addition of substances capable of rendering the colour soluble in water, and these have included the addition of sulphuric acid (as noted above), trisulphide of arsenic (the Orpiment bath) used in India (Irwin and Brett, 1970), or the introduction of sodium dithionite (first made available in 1904 under the name sodium hydrosulphite) the two latter having the effect of de-oxygenating the colour in the

dye vat, and thus maintaining its solubility in water. The cloth, when dipped and removed appears yellow-green until the atmospheric air begins to act upon it, turning it slowly towards blue-green and finally blue itself (Cannon 1994).

Fortunately for printing ink production, this process is not necessary or even desirable, since all that is required is a finely divided particle of blue pigment. In this study I propose the use of concentrated indigo stock solution made up from commercially available supplies of powdered indigo. In addition I shall be adopting, with some modification, the method of processing raw indigo plant material, suggested by Dr David Hill of Bristol University (published privately Sept 1992). The method is as follows:

1. Collect fresh leaves and stalks.
2. Pack tightly in a closed container filled with warm water and leave for two to three days in a warm place.
3. Pour off liquid into a separate container and add sodium carbonate or ammonia, and aerate.

Natural indigo continued in use into the early part of the 20th



century, when the synthetic product, developed by Bayer *et al* in Germany (over a period of 20 years and a cost of £1,000,000) gradually replaced it (Furness in Armstrong, 1924). It is interesting to note that the same author makes the following statement (in 1924) regarding natural indigo:

'In spite of the great improvements in technique, synthetic indigo may still have to face the competition of natural indigo...it is possible to foresee a reduction in the cost of production of natural indigo sufficiently great to bring the latter into full economic competition with synthetic indigotin...If only intensive research on these lines be fostered, the threat of the natural indigo industry may be delayed indefinitely.'

*Chemistry in the Twentieth Century 1924*

It is tempting to suggest that world economic conditions might still be enhanced by the reintroduction of an industry which formerly created so much commercial activity and employment.

Along with weld and madder, indigo is accepted by most authors as being of extremely good fastness to light. Padfield and Landi (1966) ascribe an approximate BS rating of 5 to 7, while

Dalby (1985) gives it a BS rating of 6 and over. Hummel, in his *British Association Reports*, section B (1893 - 98) places sulphonated indigo in class 2 (Fugitive Colours) whilst Vat Indigo Blue appears under class 5 (Very fast colours). Both Doerner (1934) and Mayer (1962) are dismissive of indigo as a colour suitable for the artists pallet, due mainly to its particular hue (which can be achieved using synthetic equivalents) and relative lack of permanence, Colours required for these purposes however, are not considered useful below BS rating 7 (Stephenson, 1989)

### Chemistry

According to Sherwood Taylor (1933) and Prescott and Ridge (1933) indigo is present in the leaves of the plant as the glucoside indican, a colourless water soluble substance, which undergoes hydrolysis with dilute acids and enzymes (present in the leaf) into glucose and indoxyl. In the presence of air indoxyl is oxidised to yield the blue pigment indigotin (indigo), which is insoluble in water, ether or alcohol (Goodwin, 1982). For the purpose of ink production, this insoluble pigment can be ground to a finely divided particulate, and thus act as a pigment capable of suspension. The Colour Index list the following constituents of indigo, Indigotin, Indirubin, Indigo

gluten and Kaempferol.

### Cultivation

*Indigofera tinctoria* as has been stated is a native of the tropics, and as such is not ideally suitable for cultivation in Britain. However some species are available, and can be successfully grown in temperate climates, notably *I. gerardiana* syn. *I. hetrantha* (Cannon, 1994) and *I. potaniii*, where they require a sunny position, in good, well drained soil (*Readers Digest Encyclopaedia of Garden Plants and Flowers*). The latter reference states that they should be planted out in either September/October or in April. The crop should be harvested as the plant begins to flower, when it is cut down to just above the root. In ideal conditions, up to four croppings can be obtained in one year, though this would not be possible in temperate climates (Rhind, 1872). I have also seen *Indigofera pseudotinctoria* on sale in garden centres for outside cultivation, and Dr John Wilkinson, of Middlesex Universities Herbal medicine Department, suggests, in a private communication, that other species may also be grown in this country. The RHS Plant Finder (Philip, 1996) list seven species of *Indigofera* available for cultivation in Britain, some of which are viable only under glass. Of those species belonging to other families, but which are capable of producing indigo blue pigments, none appear to be

none appear to be recommended in the literature as viable in Britain, with the exception of *Nerium tinctorium* syn. *N. oleander* , *Mercurialis anna* and of course the native Woad (*Isatis tinctoria* ) which will be dealt with later. Indigo is today, as in times past, largely imported from india.

### Nomenclature

The colour derived from the main Indigofera species has been known by the following names; in France, Anil; Spanish, Anil o indigo; India, Neeli ; Zimbabwe (Shona) Mutsanana, Mutsvairo. In addition, the sulphonated version of indigo is often called Saxon Blue.

### Woad (*Isatis tinctoria*L. Cruciferae)

Woad is described as a biennial herb of the genus Cruciferae (the cabbage family), which according to Goodwin (1982) is indigenous to Assyria, western Asia and the countries surrounding the Mediterranean, but that it has been grown as far north as Scandinavia since historic times. Leggett, writing in the Journal of the New York Botanical Garden. (Vol 45 [533] pp114-118, 1944) claims Southern Europe as its origin. It forms a rosette of toothed, oblong or obovate leaves from which arise stems, branching near the top, and on which

leaves from which arise stems, branching near the top, and on which are formed lanceolate to linear glaucous sessile leaves, reaching a height of around five feet. The flowers are numerous, small and born on paniced racemes formed in early to mid summer, followed by pendulous black seeds (Stuart, 1979). Cannon (1994) describes Woad as biennial or perennial, forming a rosette of leaves in the first season, followed by an erect stem the following year. Rhind (1872) describes the plant as being loaded with thick leaves 'of a lucid green colour...about a foot long and half a foot broad at their widest part.'

Perkin and Everest (1918) record the use of cabbage (*Brassica oleracea*) and Rhubarb (*Rheum rhabonicum*) to produce what they refer to as 'spurious woad,' but state that it was of inferior quality.

Woad is probably known best for its use in pre-Roman Britain, as a body stain, and Caesar describes the inhabitants of Britain during this period thus;

'All Britons stain themselves with woad, which grows wild and produces a blue colour which gives them a terrible appearance in battle.'

*Commentarii de bello Gallico. Book 5*

This statement is echoed by a number of classical writers including Pliny (Historia Naturalis book 22), and Ovid (Amores book 2). Some

writers refer to the medicinal effect of smearing woad juice on the skin, and suggest that this may have been the true reason for its use (Rhind, 1872, Goodwin, 1982, Stuart 1979). Leggett also quotes *Guests Origines Celticae* in which he states that the word 'Britain' may originate from the latinised form of the Celtic word 'Brython' meaning 'painted men.'

As a dyestuff, woad appears to have been in use since prehistoric times. According to Cardon (1990) excavations in neolithic sites in France show evidence of the use of woad. Cannon (1994) also suggests its use in ancient Egypt. By the fifth century, the Saxon invaders of Britain used so much woad that the domestic crop was unable to satisfy their needs, prompting them to import large quantities from their former homeland (Leggett, 1944). The same author notes documents pertaining to duties payable on woad appearing in 13th century England. He further states that prior to the introduction of Indigo, woad was the only source of blue dye available in Western Europe.

The gradual rise of Indigo, a much stronger source of blue, over the period of almost 300 years from its introduction in the early 16th century, signalled the demise of woad as an item of commerce, though the last commercial cultivation in Britain ended when

Skirlbeck Mills, in Lincolnshire, the last woad mill in the world, closed after the 1932 harvest (Wickens, 1983). The chief centres of woad production in Britain were in Somerset, Lincolnshire and East Anglia (Goodwin, 1982), but the most important woad growing areas in Europe as a whole, were in Saxony and Thuringia, in northern and central Germany, from where the product was exported to many parts of Europe including Flanders, England and France (Leggett, 1942). The *Journeys of Celia Fiennes* written at the end of the 17th century, contains a very useful description of woad cultivation and production in Toddington (Gloucestershire):

'..all the summer season if drie for four or five months they sow it...when grown up a little out of the ground for it rises no higher than lettice, and much in such tuffts... this they cutt off close to the ground and soe out of the same roote springs the same leafe againe; this they do four tymes, then in a Mill with a horse they grind the Leaves into a paste, so make it up in balls and drye them in a Penthouse, to secure it from raine, only the wind dryes it; this plantation of about 12 acres would employ 2 or 3 familyes men women and children... the smell of the Woade is so strong and offensive you can scarce beare it at the Mill: I could not forse my horse near it.'

*The Journeys of Celia Fiennes c. 1694*

Duke and Edlin-White (1993), give further historical accounts of woad culture in mediaeval England.

The processing of woad into a usable blue dye or pigment follows similar lines to the processing of indigo by the fermentation method, and, as has already been stated (under Indigo) the pigment contained in the plant is practically identical. Berthollet (1792 - 1818), Bancroft (1794), Partridge (1823), Crookes (1872), Rhind (1872) all give descriptions of the fermentation method as applied to woad, varying only in minor detail. The more recent literature on craft dyeing outlines similar techniques, which can be summarised as follows:

1. The fresh leaves are washed, and quickly dried.
2. The dry leaves are ground to a smooth paste which is compressed in heaps and left for a period of two weeks.
3. Portions of this mass is moulded into balls and when completely dry, crushed into a powder.
4. Moisture is added to the powdered woad and fermentation is induced over a period of about 1 to 2 days.



5. The powder is diluted with boiling water to which is added a proportion of slaked lime (calcium hydroxide) and a fresh fermentation begins.

6. The resultant liquid dyes cloth or yarn a green colour, which, as with indigo, turns blue on exposure to air.

*Rhind's Vegetable Kingdom, 1872*

The above complicated and time consuming procedure is subject to many more refinements in practice, and not the method by which colour will be derived in this study. Again, as with indigo, other methods employing acids or de-oxidising agents are described by many authors (Cannon,1994; Goodwin,1982; Duke and Edlin-White, 1993; Dalby,1985; Goodwin,1982; Wickens,1983).

The method outlined by Dr. Hill (1992) is both quicker and simpler, in addition it requires the addition of sodium carbonate only, a far less caustic material than that which is recommended by others. A concentrated extract can also be prepared from this process and this will be the method (with possible modifications) that I propose to adopt in this study:

1. Fresh leaves are added to a container of boiling water.

2. The resultant liquid is strained into a separate vessel.
3. Sodium carbonate added turning the brown liquor to a deep green, which is then aerated to produce a blue dye.
4. A blue pigment (indigotin) is formed by precipitation, and the supernatant liquid is poured off. Fresh water is added to wash the indigotin and subsequently poured off again. Preservative (eg. sodium metabisulphate) is added and the concentrate stored for later use. The addition of Hydrochloric acid at the precipitation stage is recommended, though not held to be necessary.

Research into the commercial potential of a reintroduction of woad as a source of blue dye is currently being carried out by Cook and Stoker at the Long Ashton Research Station, Bristol. A number of varieties of *Isatis tinctoria* have been the subject of cultivation trials, and they have also succeeded in extracting the colour in a purer form, using a technique similar to that outlined above. Whilst their work is not directly concerned with ink production, their results, which also include tests of weld and madder, will be of great interest when published.

Light fastness tests for woad do not generally appear in the literature since its use, independent of other colourants, seems to have been discontinued before more scientific light testing procedures were introduced at the end of the 19th century. I can find only one reference to the lightfastness of woad (Dalby,1985) which states that a very light blue obtained from woad has a BS equivalent of only four, though she goes on to remark that higher ratings can be achieved by increasing the quantity of plant material used. However, it may be assumed that the standards recorded for indigo bearing species (see under indigo), apply equally to blues of similar intensity obtained from woad, being essentially chemically identical pigments. Light fastness tests, specifically for woad will be carried out in this study.

### Chemistry

Perkin and Everest (1918) maintained that the colouring principal of woad was distinct from that obtained from the indigoferas, though at that time the substance had not been isolated. Cannon (1994) identifies the pigment *isatan B*, as being present in woad which is hydrolysed into *indoxyl*, two molecules of which combine to form the pigment indigotin (when oxidised), which is present in the indigoferas. He adds that, as with indigo, indirubin is

also present.

### Cultivation

According to most authors (Goodwin,1982; Wickens,1983; Cannon,1994; *et al* ) woad may be sown at any time when the soil is warm, ie. between March and October. Stuart (1979), along with older references state that the soil should be very rich in nutrients (Rhind 1872), while Berthollet (1792) adds more specifically that it should be a 'good dark mould, light, and well improved' by 'double tillage.' Ponting (1980) quotes Youngs reference, in his *Annals of Agriculture* (1799), to the plants tendency to exhaust the soil very quickly. Cannon (1994) along with others, states that three or four crops may be obtained each season by taking the largest leaves, whilst allowing four or five young leaves to remain, thereby ensuring further crops within a few weeks.

### Nomenclature

Other names for woad include Dyer's Weed, Pastel (Sp.) Guede (Fr.) der (Faber) waid (Ger.) Guado ( Dutch). The ancient Greeks knew it as Isatis, hence its modern generic name.

Garden Huckleberry (*Solanum guineense* syn. *S. intrusum*) Solanaceae

Huckleberry is a member of the genus solanaceae comprising (according to various authorities) some 1400 to 1700 separate species, amongst which can be found the potato (*Solanum tuberosum*) and the nightshades (*Solanum dulcamara* and *S. nigrum*) (Oxford Dictionary of Botany, 1985). It is occasionally described as a variety of *S. nigrum* known as 'guineese', being a somewhat larger plant, and unlike its relative, producing larger, non-poisonous edible berries (Unwins Seeds Ltd 1996). It grows to a height of 2 to 3<sup>1</sup>/<sub>2</sub> feet and does not require support. Uphof (1959) places its origin in Central America, where, because of the saponaceous qualities of its fruit, it is used by the natives for washing, Zumbuhl (1986) refers also to the use of the berries of *Ccormis* (*Solanum hispidum*), as a surfactant by the wool dyers of Peru. It has been used in the United States to make pie-fillings, jams and wine and is also added to ice cream, where it is said to impart 'a lovely rich colour' (Timberlake and Bridle in Markakis, 1982; Unwins Seeds Ltd, 1996).

Uphof's exhaustive work *A Dictionary of Economic Plants* includes no member of the genus in its review of dye plants (Uphof,

1959).

The work done over the past 30 years on this plant and relating to its potential as a food colourant is the only source of information which seems to be available. No mention of its use is made in any of the standard works on plant dyes, nor is it referred to in any of the craft dyeing manuals that I have been able to consult. Its presence here in this study is based upon the the published food colour work (see below), and on my own initial tests (Shaw, 1995), which have demonstrated a quite startling array of colours obtainable from the plant, including a blue, which if it can be made permanent should prove superior to indigo or woad for colour process work.

Whilst colour is readily released in hot water, the addition of SO<sub>2</sub> at a concentration of 2000 to 3000 ppm has long been used in the preparation of a product known as 'enocianina.' This product has been prepared from grape skins for more than 100 years and is used to colour wine, it is said to increase the colour yield several-fold (Garoglio,1965). Similar observations are made by others (Peterson and Jaffe, 1969; Palmadis and Makarkis,1975; Shewfeldt and Ahmed, 1978) with reference to a range of anthocyanin bearing fruits of which Garden Huckleberry is one. Though this information comes from food colour industry, it would seem useful to examine its

application in the present study.

There appear to be no specific references to the colours behaviour in light in a dry, pigmental state, though the stability of anthocyanin pigments in general are known to be poor. Photo stability in food products, is said to be improved by the introduction of isoflavone or aurone pigments (Sweeny, Wilkinson and Iacobucci, 1981). It may be possible therefore to combine known isoflavone and aurone bearing plant material (eg. *Genista tinctoria*, *Cystisus scoparius*, *Coreopsis tinctoria*) with any of the anthocyanin producing berries to achieve improved lightfastness. It is however noted by the same authors that such an introduction causes a bathochromic shift (a colour shift towards the red end of the spectrum), and would tend therefore, to preclude their use when the desired result is blue. It would on the other hand have potential advantages in the production of magentas from pokeweed and elderberries for instance.

Again, with reference to the anthocyanin pigments in general, researchers in the food colour industry have noted the detrimental effect of high temperatures during the processing of fruit products, the general consensus being that anthocyanin pigments are readily destroyed by heat (Markakis, 1974). This observation has obvious

implications for this present study.

### Chemistry

Garden Huckleberry has been investigated by Saito *et al* (1965) and Francis and Harborne (1966) whose purpose was to investigate the plants potential as a food colourant. The latter showed it to contain 93% petanin with small quantities of negretin along with a pigment specific to Huckleberry which they termed 'guineesin'. Bobbio, Bobbio and De Souza (1987) indicate high concentrations of the anthocyanin pigment malvidin in the berries of *Solanum americanum*, which they suggest could have similar applications in the food colour industry.

### Cultivation.

According to Unwins Seeds Ltd, the seeds should be sown in good quality compost, under glass at a temperature of between 18 and 21<sup>o</sup> C. around March or April. The seedlings should be transplanted 2ft apart, after being gradually accustomed to outside conditions, in late May or early June. The berries ripen in August and September, and should be picked when they are quite black.

### Nomenclature



I can find no references to Huckleberry by any other name.

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## CHAPTER V

### Blacks

## Oak Galls

A variety of blacks have traditionally been prepared using vegetable sources, particularly from those containing significant amounts of hydrolysable tannins. One of the most ancient of these being prepared from oak galls (an excrescence caused by the puncture of a species of wasp (*Cynips gallæ tinctoriæ*) and found on the young shoots of several species of oak, most notably *Quercus infectoria* (Rhind, 1872). Oak galls are a rich source of tannic acid, the principal colourant contained within them, it can range from 25% to 75% of the total weight of a particular sample, depending on the source; the so-called Aleppo (Persian and Syrian) galls being considered the best (Rawson, Gardner and Laycock, 1926). Carvalho (1904) describes the use of galls in ancient Egypt as cosmetics, and Pliny in the first century AD speaks of their use with iron as a means of blackening paper (Mayer, 1961).

Hebrew writers in the 6th century AD are said to have used these so-called iron gallo-tannate inks (Carvalho, 1904). Syriac monks working in the 9th and 10th centuries AD, recorded recipes for the manufacture of black ink which involved boiling the galls in dark red wine or vinegar, instead of water, combining the solution with iron, and adding gum arabic as a binder. One such recipe reads as follows:

' Take  $\frac{2}{3}$  oz. of gall-nut, pound it, and put it into a new pot. Pour on it a quart and a half of water, and boil it till one-third evaporates, then strain it through a course cloth, and put again into the pot upon the fire. Take  $\frac{1}{3}$  oz. of gum-arabic, and pound it till it becomes like dust. Let the water cool; then take the gum-arabic and through it into it by little and little. Take an ounce of copperas; pound it very fine and through it in . Then put it (the pot) on ( the fire) again, till it (the water).....(?). Then take it off, put it into a bottle, and let it stand for three days.'

W.H.P. Hatch 'An Album of Dated Syriac Manuscripts' Vol III <sup>1</sup>

Theophilus, writing in the 12th century AD gives similar recipes for their preparation (Theophilus *trans.* Dodwell, 1961). The introduction of fish glue (isinglass) mentioned in the 'Secretas' of the 12th century AD., is said to 'render it more impervious to the natural elements or of chemical reagents' (Carvalho,1904). Indeed research now being undertaken at the British Museum's Department of Conservation suggest that isinglass does, in some way, inhibit the fading of orpiment (arsenic trisulphide), a mineral pigment much

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<sup>1</sup> British Museum MSS: Add. MS.14540 (No. DCXXVIII),fol. 54 (marg); Add. MS 14632 (No. DCXCCV), fol. 2; Add. MS. 14644 (No. DCCCCXXXVI), fol 94; and Arundel Orient . MS. 53 (Rosen and Forshall, No. LXII), fol 83v.



used in antiquity (Lee,1995). Her belief that isinglass is not likely to exert the same protective effect on vegetable sourced colourants, may not be altogether justified, in the light of these medieval claims and will be subject to further investigation. Carvalho (1904) cites a number of manufacturers who were producing quantities of high quality writing ink made from galls and ferrous compounds as recently as the 1880's.

Textile Dyers have long used oak galls, sometimes referred to as nut galls, to produce blacks on cloth, and also as mordants to fix other colours. Reference to this is made by a number of authors (Bancroft, 1813; Berthollet, 1823; Partridge, 1823; Crookes, 1872 *et al*) and more recently a number of recipes can be found in the craft dyeing literature (Adrosko, 1972; McGrath, 1977; Duke and Edlin-White, 1993).

The high tannin content of many barks (eg. oak, sumac etc.), nuts (eg. black walnut hulls, chestnuts shells etc) and leaves (alder,birch and beech) are said to produce similar blacks when combined with ferrous materials but are often described as dark grey, rather than true blacks ( Lesch, 1970; McGrath, 1977; Ponting,1980; Cannon, 1994). My own research has succeeded in producing an

extremely dense black by combining, in boiling water, horse chestnut shells, ferrous sulphate and sodium carbonate (see Appendix 3).

With the exception of black walnut hulls, none of the above ever achieved the status of commercial dyestuffs, on a par with oak galls. Galls however, by their very nature, are less capable of controlled cultivation than the true plant sources.

Logwood (*Haematoxylon campechianum*) L. Leguminosae

The arrival in Europe of Logwood (*Haematoxylon campechianum*) in the 16th century was to satisfy the need for a cheap, reliable and versatile source of black for the dyeing industry, right up until the present day. It is one of the few vegetable colourants capable of dyeing synthetic textiles. (Fox, 1987).

Logwood is a large leguminous and rapidly growing tree cultivated in South America and the Caribbean, where Rhind (1872) tells us crops became established within a few years of planting. It is a small, crookedly-branched tree, the branches being spiny and the bark rough and dark. The small heart shaped leaves have four pairs of leaflets with the points towards the stem. The flowers are small and yellow, with five petals, growing in axillary racemes.

It appears commercially in the form of chips, rasped or ground powder and in the form of a liquid or solid extract ((Rawson,Gardner and Laycock, 1924). Its colour principal, haematin, is altogether distinct from that of oak galls (Perkin and Everest, 1916; Mayer, 1943). The literature abounds with recipes for blacks using logwood, which is also capable of producing less light fast blues and purples - all the sources thus far mentioned in this review provide recipes for this dye.

As with oak galls the use of ferrous material is essential for good blacks, though potassium dichromate was eventually to become the more common mordant after its introduction, possibly in the early part of the 19th century (Ponting, 1980).

A process known as 'aging' or curing' the raw material was long considered essential to fully develop its dyeing potential. A typical method of 19th century preparation required several weeks to complete, involving the constant observation and turning of layers of dampened logwood chips, the object of which was to maximise the haematin content by oxidisation. The so-called French and American processes (the latter involving extraction under pressure, but both requiring the introduction of some oxidising agent such as hydrogen peroxide) introduced in the latter part of the 19th century, which

provided the industry with dyes of equal intensity from freshly cut material, demonstrated the redundant nature of the aging procedure (Rawson, Gardner and Laycock, 1924). This sequence of events is reminiscent of a number of other complicated procedures involving other dye materials which were subsequently superseded by speedier and infinitely less labour intensive modes of manufacture (see indigo and woad).

Logwood is still grown for export on the island Haiti and can be obtained in Britain both as raw chips and dried extract (Dean, 1996). It remains - despite low commercial consumption - one of the cheaper natural dye sources.

### Other Sources

For the purposes of this study, many of the so-called blacks mentioned in the literature are far from ideal since they might more accurately be described as dark blue or red. Indigo and madder were often 'top dyed' with logwood to achieve extremely dark blues and reds, which, to all intents and purposes could be sold as blacks. All of Partridge's black recipes for instance, involve the use of logwood with other colourants (Partridge, 1823), as do many of those

described by Bancroft, Berthollet and Crookes. The problem with regard to the production of 'process' blacks is that any hint of red or blue in the black ink would only tend to reinforce the magenta and cyan constituents of the colour separation work, rather than providing the necessary tonality that a true black should provide. The black used for the purposes of this study should therefore be as chromatically neutral as possible, and a logwood/ferrous combination would appear to be one of the most promising sources with which to proceed. In light of the results I have obtained producing magentas and blacks from elderberry, it should be mentioned that others have achieved similar results from the same source. McGrath (1977) and Cannon (1994) each describe blacks achieved with elderberries and ferrous sulphate.

As far back as the 1st century AD, Pliny writing in his *Historia Naturalis* (Book XXXV 25, 42.) speaks of a black ink perfected by the painters Polygnotus and Micon, of Athens, using grape skins, to which they gave the name (according to one translation) Trygynon, but gives no details as to its manufacture.

In addition several authors describe a number of other sources. Dalby (1982) cites Iris roots (*Iris pseudocorus*) mordanted with iron. Lesch (1972) describes the use of Barberry (*Berberis sp.*), Cutch

(chiefly *Mimosa sp.*, *Acacia sp.* and *Areca sp.*) and multiflora rose leaves and shoots, again in combination with iron, in the form of ferrous sulphate. Cannon (1995) recommends the rhizomes of water-lily (*Nymphaea alba*), Weld (*Reseda luteola*), or heather (*Calluna vulgaris*), each with potassium dichromate and iron, and the leaves of black walnut (*Juglans nigra*) combined with alum and copper or ferrous sulphate.

Adrosko (1971) refers to the use of a variety of plants, used in North America during the 18th and 19th centuries, though not always detailing their preparation, they include the following:

Poison oak (*Rhus toxicodendron*)

Gypsywort (*Lycopus europaeus*)

Baneberries or Herb Christopher (*Actea spicata*) with alum

Malacca bean (*Semecarpus anacardium*)

Sorrel (*Rumex acetosella*)

*Andromeda arborea*

### Lightfastness

Generally speaking, all colours mordanted with ferrous compounds have a high resistance to the effects of light. Both the logwood and oak gall blacks are usually described in the literature as reasonably fast to light. Hummel (1898) places logwood black in class

reasonably fast to light. Hummel (1898) places logwood black in class III, of his five categories, ie. 'moderately fast to light.' Padfield and Landi (1966) gives a BS equivalent of 3 for logwood mordanted with iron, Dalby (1985) gives an identical rating for iron, and 4 with chrome. The Colour Index describes a variety of logwood applications on silk, wool cotton etc as being, 'good', 'very good' and 'very good to excellent' with regard to lightfastness. It appears in the CI as Natural Black 1,2,3 and 4 (CI 75290 and CI 75291).

#### Nomenclature - Oak Galls

Oak galls have been referred to as, nut galls and gall-nuts, Aleppo Galls, *Noix de Galle* (Fr.) and *Galapfel* (Gr.).

#### Nomenclature - Logwood

Logwood has been variously described as, blackwood, bluewood, bloodwood, campeachy wood, Noir direct, Peachwood. Bois de Campechey de Sang or d'Inde.

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## CHAPTER VI

### Thickening Agents, Preservatives and Other Additives

In order to prepare coloured printing inks from aqueous plant extracts suitable thickening agents must be introduced to the liquid colour. The inks viscosity must be increased to the point at which it remains stable, both in the screen, and on the printed substrate, whilst at the same time passing through the screen mesh easily without clogging (Taussig, 1950). Nicholson (1993) referring to pigment suspensions sums up these qualities as follows:

While wet, the coating must be capable of being spread evenly across the surface and of remaining spread without developing faults, such as crawling or pinholing. The film must solidify in such a way that it does not pull itself off the substrate; and once solid, the film must maintain integrity as a film. It must not, for example, form a discontinuous powder. It must also retain adhesion to the substrate in the solid state.

Referring to the definition given by Sanders (1989), the coloured lakes produced in this study may be considered as pigment suspensions, while those inks formed by a simple thickening of a coloured aqueous extract may be regarded as thickened dye solutions. The principles outlined above however remains true for inks of either formulation, since the thickener remains as an integral part of the

coating throughout its life.

Whilst screen process printing is a relatively modern technique, the first patent (BP No 756 ) being granted to Samuel Simon, a Manchester artist and designer, in 1907, the history of thickened inks for various 'block printing' techniques is a long one. In addition, the 20th century textile screen printing industry provides much valuable information on the subject.

Many of the thickeners referred to in the literature covering block printing, and spanning some 4,500 years of history (Story,1974), can still be seen in use in today's water-based textile screen printing operations, along with a number of newer synthetic preparations. In this study, I propose to use vegetable thickenings exclusively.

Many vegetable gums and starches have been used as water soluble thickening agents. In an article on thickeners (CIBA Review 107), the author lists the following gums and starches in common use during the 1950's : Tragacanth (*Astragalus sp.*), wheat (*Triticum sp.*), maize (*Zea sp.*), potato starch (*Solanum tuberosum*), British Gum (heated wheat starch), gum arabic (*Acacia senegal* and other species), locust or carob bean flour(*Ceratonia siliqua* ) crystal gum, alginates (*Macrocystis sp.* and others) and cellulose derivatives. Other authors

(Crookes,1872; Rhind, 1872; Doerner,1934; Mayer,1962; Stephenson 1989) site the following: gommeline, gomma (both varieties of dextrine), bassora gum (a tragacanth constituent), salep,(*Jatropha manihot*), sago (*Cycas sp.*), linseed (*Linum sp.*), gum karaya (*Sterculia urens* ), cherry gum (*Prunus sp.*), rice starch (*Oryza sp.*), sarcocolla (*Astragalus sarcocolla* ) and arrowroot (*Maranta arundinacea*). In the food processing industry, a similar range of vegetable thickeners can be found, some of which are, or may be of use in print applications. Hanssen (1987) lists the following: sodium, potassium, ammonium and calcium alginates, agar(*Gelidium amansii*, and other members of the genus Gelidaceae, along with the geni *Sphaerococcaceae* and *Rhodophyceae*), carrageenan (*Chondrus sp.*), guar gum (*Cyamopsis teragonolobus* and *C. psoraloides*) and corn sugar gum (*Zea sp.*).

It is proposed that of the thickening agents which have traditionally been used in the textile printing industry, gum arabic, potato starch and the seaweed derived alginates will form the basis of the thickeners used in this study. As with the dyes themselves, thickeners for textile applications have differing, requirements to those suitable for the purposes of this study. In general terms, printed paper applications are less demanding. There is, for instance, no need

for the thickener to be subsequently removed from the substrate, as it is in textile printing (by washing), nor is it necessary to take care not to use combinations of thickener which may damage the fine filaments of certain synthetic fibres. Never the less, many of the considerations which must be taken into account for textile use, are equally important to the proposed application, and these are outlined below.

### Starches

Mara (1979) suggests the use of cellulose wallpaper paste as a thickener for water-based screen process inks, but excluding this, processed potato starch, available in Britain under the trade name *Printex*, is probably the cheapest commercial source of thickener. It can be used alone, but better results are obtained when a proportion of vegetable oil, glycol, honey, molasses or glucose is added to inhibit drying and thus improve the mixtures flow characteristics (Middleton, 1960). The CIBA Review(27) states that starch reacts with a number of substances, rendering it useless for print applications. Starch hydrolyses in combination with mineral acids and their metal salts, eg. aluminium sulphate, they become thin and unusable, an effect long appreciated by others including Crookes (1872). The contrary effect is noted when starch is combined with

colour which is too alkaline resulting, at worst, in a gelatinous substance known as Aparitin. It is clear that it would be safer to achieve a fairly neutral pH value for colours thickened with potato and other starches.

Crookes goes on to say that the acetic salts however, formed from aluminium, iron and chromium, which are known to form lakes with plant colourants, will combine with starch to form compounds which, though difficult to remove from textile fibres, may prove useful, for that very reason, in the production of water resistant inks of the type proposed here. The various farinaceous starches have one other effect besides their ability to thicken aqueous plant extracts. According to Crookes (1872), they also prevent oxidation of the printed colours, which, in most cases is an added advantage. He adds that inks thickened with dextrine, a product derived from starch, tend not to keep as well as those thickened with gums.

Effective preservatives for starch thickened inks include the commercial preparation *Preventol* or alternatively a solution of 0.5% formaldehyde (Mayer, 1962). Stephenson (1989) recommends halogenated or chlorinated phenols, boric acid, sodium benzoate, or, if it is present in high enough concentrations, alcohol.

## Gums

Taussig (1950) regards gum arabic as superior to other gums when reproducing detailed work, though the addition of 20 to 30 gm/ltr of olive oil (providing the colour is not too alkaline) is necessary to achieve a less brittle finish. Gum tragacanth is a more expensive source (as are the gums karaya and locust bean), though it is more flexible, and has a thickening capability some ten times that of gum arabic (Stephenson, 1989). Dean (1993 and 1995), recommends the use of gum arabic alone, stating a proportion of 100 gm of gum to 100 ml of water, though this would seem too thin for screen printing applications.

As with the starches, gum thickeners benefit from the addition of plasticisers, such as glucose, glycerol, honey, molasses etc (Middleton, 1960; Mayer, 1962; Stephenson, 1989). Inks thickened with gum, are generally more resistant to attack by micro-organisms, but should never the less be protected by the addition of anti-bacterial preparations eg. 0.5 to 1% sodium orthophenyl phenate, 0.5% beta naphthol (Mayer, 1962). (cf. 'Starches')

## Alginates

Although the potential of seaweed to produce thickening agents



has long been known, an effective industrial process was not developed until the 1930's, production in Britain and the USA was finally realised after 1945 (CIBA Review 1969, 1). The same source states that the chief product used as a thickener in print applications is the sodium salt, sodium alginate, which it says compares very favourably with other thickeners, providing 'several practical advantages.' Among these are its ready solubility in cold water, its high viscosity and its compatibility with a large range of dye and pigment solutions.

Dean (1995) also recommends sodium alginate, in the form of the commercial product *Manutex* though not for use with mordant printing. This last remark, probably alludes to the problem of gel formation when in the presence of strong alkaline solutions (above pH11), or chromium, zinc or iron salts (CIBA Review, 1969,1) none of which, with the possible exception of ferrous sulphate, are proposed in this study. As with other vegetable thickeners, sodium alginate requires protection from fungal and microbial attack, the use of preservatives is once again essential. (see 'Preservatives').

### Starch /Gum / Alginate / Combinations

Taussig (1950) has much to say regarding thickeners for textile

printing, the difficulty being, to try to separate those merits which are of use in this present application, from those that are not. Referring to the relative anti-clogging properties of various materials, he states that, '.. a mixture of gum tragacanth and British gum (heated wheat starch) is a compromise which has been found successful.' This view is held too by the CIBA Review 27 which states that starch-tragacanth combinations are 'extremely useful' though it points out the gel forming deficiencies of this thickening in the presence of caustic alkalis, (a problem it shares with sodium alginate) noting, in addition, that even in the presence of quite mild alkaline carbonates, tragacanth can also take on an intense golden yellow colour, which can transfer itself to the substrate. Interestingly, Taussig also points out that certain starch-tragacanth combinations are not recommended for textile applications because of the difficulty of washing off after printing. As has been noted already, this procedure is not required for printed paper applications, and starch-tragacanth combinations may therefore be considered as a possible thickenings for our purposes. Crookes (1872) does not recommend the combination of gum arabic with any of the then known thickeners. Franken, writing in the CIBA Review 107, suggests a useful combination of wheat starch and alginate to overcome what he describes as a reduced colour yield when alginates

are used alone. He also suggests the use of salicylic acid (which is found in the leaves of willows and poplars) as a preservative for alginate thickeners.

### Vegetable Preservatives

Hayward (1990) outlines a number of disadvantages to water-borne coatings, including the tendency to offset their environmental advantages once biocides are introduced. In keeping therefore, with the non-toxic focus of this investigation, it would seem worth while to review some of the many permitted preservatives used in the food processing industry. Hanssen (1984) lists a number of preservatives which have the added advantage of being vegetable in origin. Sorbic acid (E200) which occurs naturally in the fruits of mountain ash (*Sorbus aucuparia*), potassium sorbate (E202, a neutralised form of sorbic acid) and benzoic acid (E210), which is chemically synthesised, but occurs naturally in the fruits of the elder (*Sambucus nigra*). The use of salicylic acid, derived from vegetable sources has already been noted.

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## CHAPTER VII

### Materials and Methods

The plants used in the initial tests were obtained from a variety of sources. Some were collected from the wild or from domestic and public gardens, others from commercial suppliers. Much of the commercially obtained material had its origin overseas (including those species which are viable in, and in some cases native to the British Isles). The individual source (where known) of each plant tested is noted in the list of initial tests (appendix 2).

### The Ink Garden

Before going on to describe the test procedures, one particular source of plant material should be mentioned. Whilst commercially obtained material is often extremely reliable and colour consistent, its exact origin and purity, along with the particular conditions under which it was grown, is impossible to ascertain. Historically, madder was sometimes adulterated with brick dust for instance. It is known that whilst some plants may be stored for quite long periods of time, with no apparent change in the colour obtained, others can deteriorate with time. For reliable results therefore it is important that samples are obtained for testing, where the origin, time of planting and growing conditions are known. It became necessary therefore, to establish a test plot in the UK. In November 1994 an area of ground

was marked out on the Cat Hill campus of Middlesex University measuring some 35 feet by 25 feet (Figs.4 to 7). The plot was ready for planting out in April of the following year. During the first year, 19 species were planted, several of which were subsequently discarded. Of these, *Arctostaphylos uva ursi*, *Prunus padus*, and *Hypericum perforatum* had to be removed due to infestation and disease. *Carthamus tinctoria* did not grow well and it too was removed. *Daphne mezereum* received the unwanted attentions of a species of deer but survived after being protected by a wire mesh cover. The discarded plants were replaced by *Isatis tinctoria* (woad) the only native source of indigotin, *Reseda luteola* (weld) *Rubia tinctoria* (madder) *Tagetes sp.* (marigold), *Anthemis tinctoria* (chamomile). In addition a number of plants were cultivated because it seemed from the literature that they may prove useful sources of blue, though rarely used. The petals of cornflower (*Centaurea cyanus*) in use during mediaeval times as a blue ink for manuscript illumination, did not provide sufficient quantities to be regarded as a serious source, nor did the particular blue obtained seem close enough to process cyan. The roots of elecampane (*Inula helenium*) a supposed source of blue, provided only a rather dull yellow. Both of these were discarded.



Fig 4. The Cat Hill Ink Garden, May 1995



Fig 5. August 1995





Fig 6. May 1996



Fig 7. August 1996

the plants need to establish themselves over a period of years, and it was therefore not possible to harvest even small quantities of material until two or even three full seasons had elapsed. Elderberry, sumac, buckthorn and mahonia, have not produced significant crops to date (autumn 1998). Fig. 8 lists the species under cultivation in September 1999.

In addition, a variety of species, some not normally capable of survival in the UK, were cultivated under protection. *Relbunium hypocarpium*, obtained from Dr Claudina Moresi of the Universidade Federal de Minas Gerais, Brasil, is a madder-like plant native to South America, containing similar pigments to madder. This species, unlike madder, contains a higher proportion of purpurin to alizarin, than Europeans cultivated madder, and thus presents the possibility of a more violet red (ie, magenta). Several species of indigofera, including *Indigofera tinctoria*, *I. amblyantha*, *I. australis* and *I. pseudotinctoria* were also sown under protection, though the latter is certainly viable in the UK. In addition to these sources of indigotin *Polygonum tinctorium*, an oriental source of indigotin was also planted. *Eleocarpus reticulatus*, a native New Zealand plant said by Uphof to provide a blue colourant, *Quercus velutina* (Black Oak) introduced as a yellow dye by Edward Bancroft in the late 18th century, and

*Caesalpinia gilliesii*, a relative of brazilwood, were also sown. Both *Quercus velutina* and *Caesalpinia gilliesii* survived the hardening off process well. *Eleocarpus*, after several attempts, failed to germinate.

### Colour Testing

Appendix 2.1 shows a range of potential process colours derived from 23 of the original 76 plant species and varieties tested, followed by the complete annotated list (appendix 2.2). The influence of 5 traditional textile dyeing mordants (modifiers)<sup>1</sup> and 5 other compounds<sup>2</sup> known to have an effect upon the extracted colour is also described, along with the temperature at which the samples were heated in water and their approximate acidity/alkalinity indicated as greater than, or less than 7.

The sources were drawn mainly from the existing literature on textile dyeing. As has already been stated, many were collected from parks and gardens, waste ground, and botanical collections  
(principally from the Chelsea Physic Garden, The Museum of Garden

<sup>1</sup> The term 'mordant' as applied in textile dyeing denotes a substance which allows the dyestuff to bond or 'fix' molecularly to the fabric or yarn. In this present study, such bonding or fixing is unnecessary, since the colour does not have to withstand the effects of washing. It is the additional effect of a mordant, ie. to cause colour change, that is of importance here. Thus it is proposed that for the purposes of this study such mordants be termed 'modifiers' since this more accurately describes their function.

<sup>2</sup> On some samples the effect of a variety of other compounds was also tested and these are listed in Appendix 2.

History, and the Royal Botanical Gardens at Kew in London), others were obtained from commercial suppliers of dye-plant material.

While many of the tests produced quite concentrated extracts of colour, most of these were considered unsuitable for the present application. It can be seen that in these tests, the most common colour found in plant material, other than the ubiquitous brown (using aqueous extraction methods), is yellow, and this is born out by the literature.

Since my interest lay in the production of process colours, many sources of purple, green and orange, whilst useful as general inks, were discounted, and a list of potential process colour progenitors was arrived at. This section deals firstly with the extraction and colour modification tests applied to these species, beginning with the procedure adopted for the initial tests, and secondly, with the more detailed tests made on those plants thought most likely to provide a range of process colours.

### Initial Tests - Extraction and Colour Modification (see Appendix 2)

The plant material was heated in mains tap-water to boiling point and simmered for periods of between 30 to 60 minutes

(depending on the denseness of the material used). Almost all references to madder processing make the observation that the red colouring matters (chiefly alizarin and purpurin) are adversely affected at temperatures exceeding 70° C. In the initial tests I followed this advice, though later it became clear that the pigments contained in the root can remain substantially unaffected by temperatures of over 100° C. provided the root is first washed thoroughly in a solution of sodium sulphate (Glauber's salts) and secondly treated with aluminium sulphate. The resultant liquor was strained off and approximately 2.5 ml was placed in each of 11 sections of an indented mixing pallet. Approximately .05 gm of one of the chemical modifiers was added to each of the 10 samples, leaving the 11th sample unmodified, the resultant colour being applied to a labelled test sheet. The chief modifiers used were as follows:

#### Potassium Aluminium Sulphate (Alum)

An ancient modifier used in textile dyeing and is perhaps most commonly known today as an astringent forming as it does the main ingredient in the so-called styptic pencils used to stem the flow of blood from minor shaving accidents.

### Ferrous sulphate

Iron has been used for centuries in combination with tannins to produce dark black/brown writing inks and the results here confirm this observation. It is interesting to note too that persian berries, a source of yellow with almost all modifiers, provides a quite dense black when combined with ferrous sulphate.

### Copper sulphate

Another traditional mordant used in dyeing, important during the 17th and 18th centuries, when it was known as blue copperas. As with potassium dichromate, its use in this study seems limited, since it appears to have no modifying effect upon the potential process colours.

### Stannous chloride

This metallic salt, introduced into the dyeing industry around the beginning of the 19th century, is said to have a brightening effect, particularly on madders, though its general effect, as seen in the test sheets could be characterised as lightening, rather than brightening.

### Potassium Dichromate

Introduced during the first half of the 19th century, potassium dichromate was to become a mainstay in textile dyeing, particularly for producing blacks on cotton. However, as can be seen in the test sheets its application in this study seems unnecessary since it provides no useful modifying effect in any of the potential process colours.

### Sodium carbonate Sodium bicarbonate and Potassium carbonate

All of these alkaline reagents produce the opposite effect to that produced by the acids like citric or acetic. Anthocyanin pigments are generally shifted towards blue/green with the addition, for instance, of small amounts of sodium carbonate. They have however another role to play which can be very important, particularly with regard to the intensity of colour released from any given sample of plant material. As can be seen from the test sheets, unmodified samples are often chromatically weaker than those which have been prepared in alkaline conditions. Though this can be a useful way of maximising the yield from a given sample, pigments which are sensitive to changes in pH are not susceptible to this method since they will also change hue as a result of the introduction of an alkaline material.

### Potassium hydrogen tartrate

Potassium hydrogen tartrate, or cream of tartar, was a frequent addition to the textile dye-bath where it was used in combination with other mordants. On its own it has a similar effect to that produced by the acids already mentioned.

### Citric acid

Though not used as a traditional mordant, citric acid (along with a number of other organic acids including acetic acid) has the effect of shifting the hue of the anthocyanin pigments found in fruits of elder, mahonia and solanum guineense etc., towards a purer red and away from the typically claret colour produced by these fruits.

At this point it became possible to assess the utility of each of the modifiers (see Fig. 8). It was obvious that some were plainly unsuitable, since they appeared to have no useful modifying effect. Others were seen to duplicate the effect of another, and some caused changes which though interesting, were not useful within the context of this study.



<i>MODIFIER</i>	<i>COMMON</i>	<i>EFFECT</i>
<b>potassium aluminum sulphate</b>	alum	brightening yellows and as a precipitate for lakes.
<b>ferrous sulphate</b>	iron	producing blacks in combination with logwood or other tannin rich materials eg. barks, galls and some fruits.
cupric sulphate	copper	often used to produce greens in textile dyeing- of little use in the context of this research.
stannous chloride	tin	brightening madder reds.
potassium dichromate	chrome	a strong yellow colourant in itself, it is used in combination with logwood to produce blacks.
<b>sodium carbonate</b> sodium bicarbonate potassium carbonate	washing soda baking soda potash	hastening cellular breakdown by softening water, thereby maximising the release of colour. The alkaline modifiers are also used to shift anthocyanin pigments from reds through blue to green
potassium hydrogen tartrate	cream of tartar	brightening yellows
citric acid		causing a chromatic shift towards the red in anthocyanin pigments

**Fig 8.** The initial tests set out to determine the effect of ten modifiers upon the extracts from 76 plant species and varieties. Cream of tartar tended to duplicate the effect of alum while citric acid, useful with anthocyanin pigments, had no significant effect on madder, weld or indigo. The use of tin, copper and chrome was considered unacceptably toxic, though chrome is said to produce the most effective logwood black. The modifiers currently being used are indicated in bold type.

It was with some relief that I was able to discontinue the use of copper sulphate, potassium dichromate and stannous chloride, since each of these are regarded by most health and safety authorities as hazardous materials. The alkaline modifiers (sodium carbonate, sodium bicarbonate and potassium carbonate) in most cases gave results so similar as to render the continued use of all three superfluous.

### The Detailed Tests

As a result of the initial round of tests, a total of 11 plants were selected for further testing, two sources of yellow, four magentas, three blues and two of black (Fig 9). These 11 sources are illustrated in appendix 3. This second round of more detailed tests sought to determine the following:

1. A more objective standard/vegetable process colour comparison, using spectral reflectance equipment as opposed to naked eye comparison.
2. A comparison of each samples resistance to light with that of standard commercial process pigments.
3. A comparison of thickened aqueous extract inks with those prepared as lakes.

<b>cyan</b>	<b>magenta</b>	<b>yellow</b>	<b>black</b>
<i>Indigofera tinctoria</i>	<i>Rubia tinctoria</i>	<i>Reseda luteola</i>	<i>Oak galls</i>
<i>Isatis tinctoria</i>	<i>Phytolacca americana</i>	<i>Rhamnus saxatilis</i>	<i>Haematoxylon campechianum</i>
<i>Solanum guineense</i>	<i>Sambucus nigra</i> <i>Caesalpinia echinata</i>		

Fig 9. The potential sources of process colour identified from the initial tests.

In the initial tests, un-thickened colours were simply brushed onto the test sheets. The second round of test samples were printed, using a 110 tpi (HD) nylon screen mesh, the colours being thickened with sodium alginate (thickened aqueous extracts). This ensured a more chromatically consistent and continuous deposit of ink, thus making the assessment of lightfastness and spectral reflectance more reliable.

The colours were extracted from between 5 to 30 gms of plant material in 250 to 500ml of tap water at 100°C. At 5 minute intervals throughout the heating process, a sample of extract was applied using a brush to Ivory card to determine the point at which further

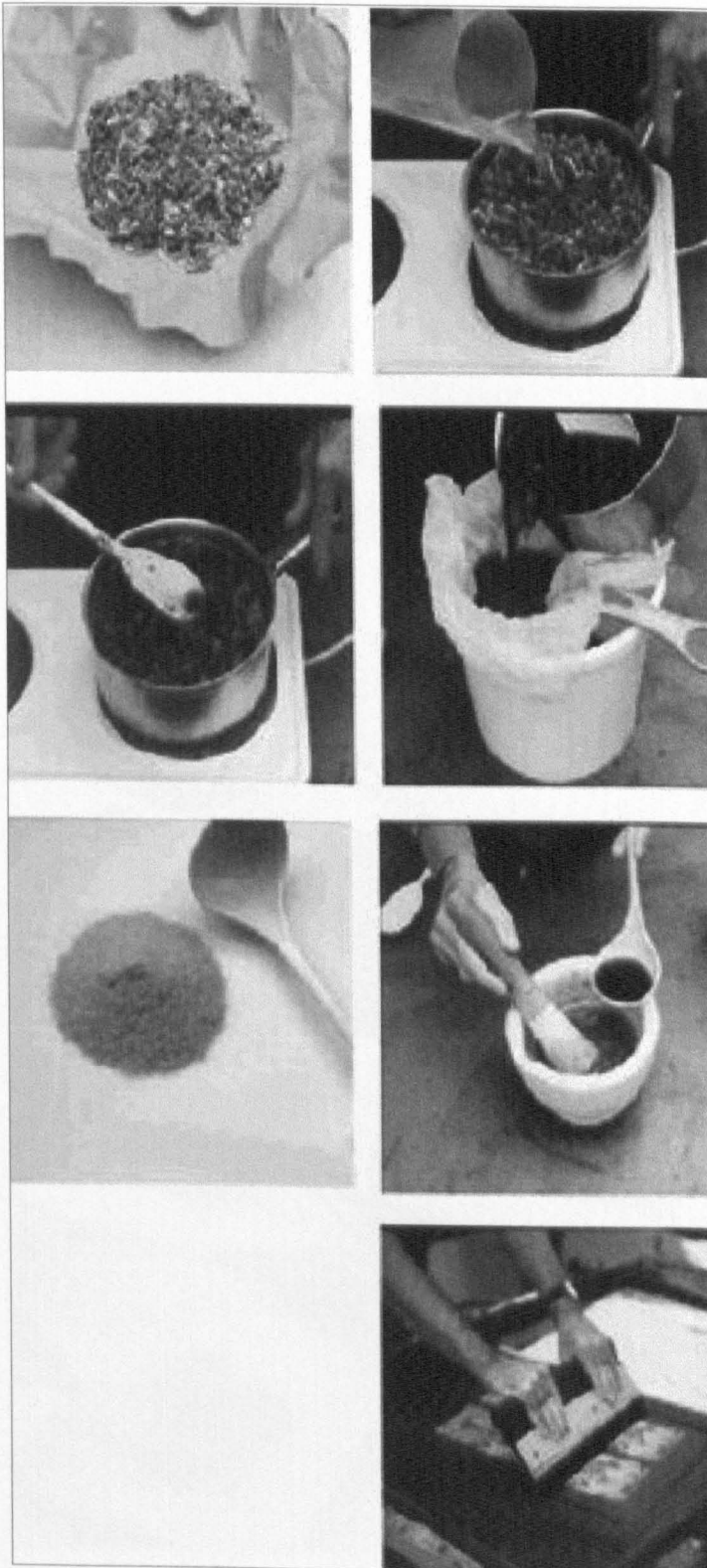
application of heat appeared unnecessary. The optimum extraction period being deemed to be that at which a samples chromatic value (its visible similarity to a process equivalent) and its transparency (the degree to which the white substrate was visible through it) appeared most closely to resemble a standard printed sample of commercial process colour. The optimum extraction period was thus found to vary for different plant sources, for most samples an extraction period of between 30 to 60 minutes at 100° C. was found to be adequate. For a given sample extracted in 500 ml of water, for 30 minutes at 100° C. approximately 300 ml. were lost through evaporation, thus leaving around 200 ml. of coloured extract. Once this optimum extraction period had been identified, the resultant colour was thickened with sodium alginate (a commercial textile dye thickening agent) and four copies of each sample were test printed onto 200 gm Ivory card. The preparation of one such thickened aqueous extract is set out in figure 10.

One set of test cards were then sent for spectral reflectance comparison tests, to determine the actual degree of similarity between the spectral curve of a given sample and its commercial process equivalent. The tests were carried out by Dr Rob Withnall of the Department of Chemistry and Life Sciences at Greenwich University,

the results are discussed in chapter 4. A second set of cards were subjected to comparative light fastness tests the results of which also appear in chapter 4.

### Lake Pigments

The coloured inks thus far described may be described as 'thickened aqueous extracts' since they are essentially dye solutions thickened to provide a printable ink. Almost all printing inks in general use (apart from those used in ink jet type printers) are pigment suspensions, rather than dye solutions. Almost all the inks used in both lithographic and screen printing, as well as artists colours and even household paints consist of finely divided insoluble particles suspended or bound in a viscous oil-based or acrylic medium. These fine particles are derived from a variety of natural organic, non-organic and synthetic sources. Vermilion, a mined pigment known in ancient times, was a natural inorganic sulphide of mercury, today it is artificially synthesised. Phthalocyanine blue a modern synthetic pigment developed in the 1930's and the source of process blue, is an organic pigment derived from copper. Chrome yellow, the Cadmium colours, Prussian and Ultramarine blue are just a few of the range of insoluble pigments used in inks and paints.



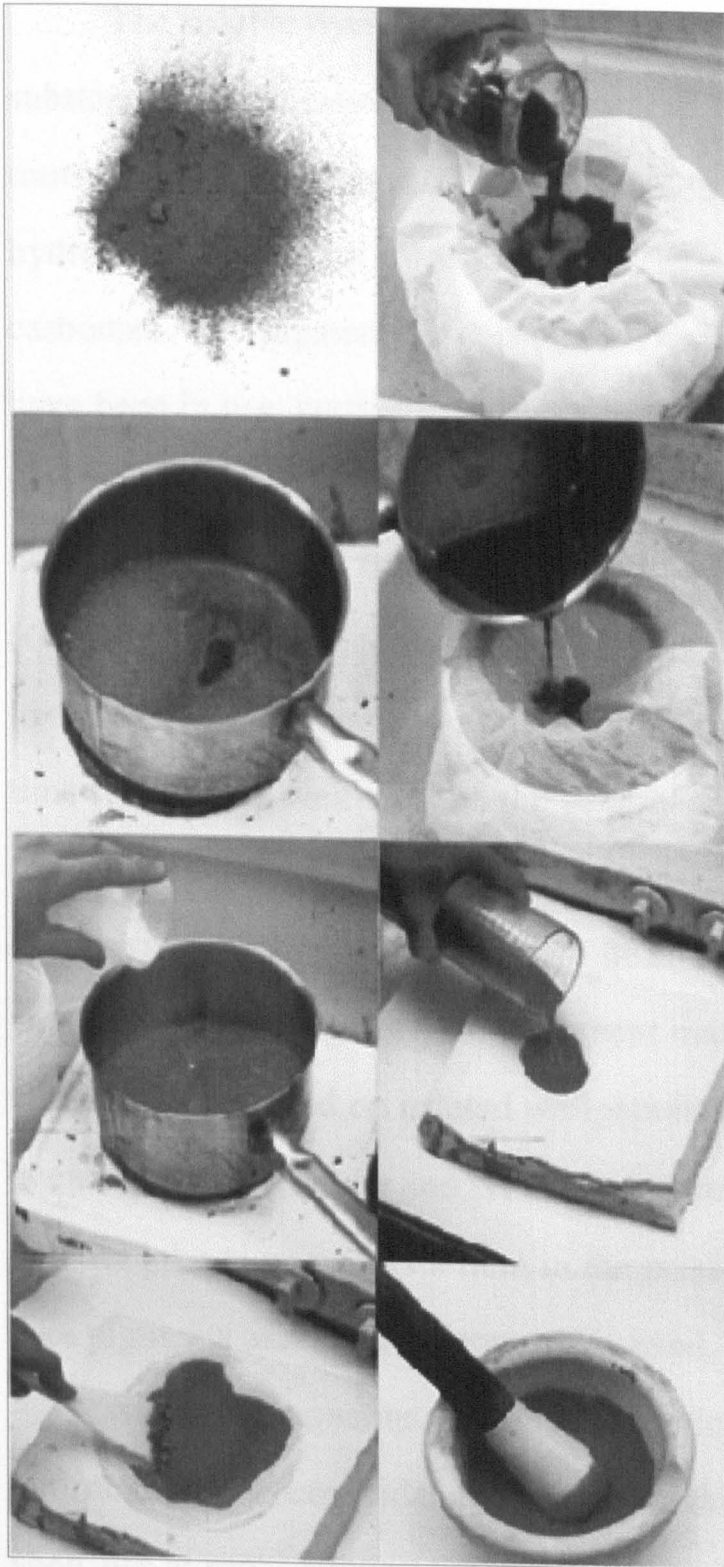
**Fig 10. Preparation of a simple aqueous extract ink from weld (*Reseda luteola*)**

20 gm of dried weld is added to 500ml of water containing 2 gm sodium carbonate and 1 gm aluminium sulphate, heated to 100° c and kept at that temperature for 30 minutes.

The coloured extract is filtered off using a finer screen mesh than will be used for printing

A thickening agent is added (sodium alginate being used here), to achieve a good printing consistency. A few drops of glycol or honey may be used as a plasticiser and an anti-bacterial/anti fungal agent such as 'Preventol' may also be added

After cooling the ink is ready to print



**Fig 11. Preparation of a lake pigment from dried madder roots.**

20 gm of ground madder root is washed in 10% solution of sodium sulphate (Glaubers salts) and filtered off.

The root is added to 500 ml of a 10% solution of aluminium sulphate and kept at 100° c for 30 mins. The extract is then filtered off.

Aluminium hydrovide is added to the extract and the whole is heated once more. After the precipitate has settled the supernatant liquid is poured off and the wet precipitate is poured onto an absorbent surface (chalk block) to remove remaining moisture.

The dried precipitate is scraped from the chalk block and ground to a smooth even consistency in a suitable printing medium

The soluble dyes from plant sources can be given similar substantial characteristics by being applied to a finely divide transparent, or near transparent substrate such as aluminium hydroxide, aluminium sulphate, barium sulphate or calcium carbonate. The pigments thus produced are known as 'lakes' and they have been in use, certainly since renaissance times. The brilliant rose magenta sleeve of one of Holbein's 'Ambassadors' painted in 1533 is a lake pigment derived from the resinous exudation of a scale insect (*Coccus lacca*) native to the Indian subcontinent. It is interesting to note that the lake pigment in this example has survived the ravages of time remarkably well, despite the general consensus regarding the poor lightfastness of lake pigments. Indeed, one wonders how many of today's synthetic colours will still be so fresh after more than 400 years exposure to light? A lake pigment made from persian berries was commonly used on printed wallpapers in the 19th century and even today the Windsor and Newton company still produce a genuine madder lake pigment. As a item in the print industry however, the lake pigments are now less commonly used.

Storage of aqueous vegetable based inks, even with the aid of antifungal and bactericidal agents is something of a problem their shelf life at room temperature being somewhat limited. Lake



pigments, on the other hand, because they exist as dry powders can be stored almost indefinitely.

A further advantage over simple thickened aqueous inks is the lakes ability to be bound or suspended in a variety of media, including vegetable oils and acrylic pastes both of which form water resistant films when dry. With these factors in mind, it seemed useful to prepare lakes of madder, weld, indigo and brazilwood to see how they compared to the thickened aqueous extracts.

### Preparation of Lake Pigments

The procedure for colour extraction is identical to that already described for thickened aqueous extracts. Once the filtered dye solution is obtained it is returned to the heat and a finely divided substrate is added (eg. aluminium hydroxide). Heat is applied until the substrate is no longer visible, at which point the solution is allowed to stand overnight at room temperature, during which time the substrate precipitates as a coloured 'sludge' with a paler 'supernatant liquor' above it. This supernatant liquor is then carefully poured off, and the 'sludge' is placed on an absorbent block (eg. Plaster of Paris or chalk) which readily draws out the remaining liquid. When drying is complete, the lake pigment can be gently scraped from the block,

ground to a fine powder and suspended in a transparent printing medium. In the print tests, the lake pigments were suspended in Rowney System 3 acrylic printing medium.

Using a modified 19th century process (quoted in *Rawson Gardner and Laycock*, 1916 and set out in fig.11) I was able to obtain a successful rose-red lake from madder root and this despite applying temperatures of 100° C. Washing the root before any heat is applied in a cold solution of sodium sulphate releases copious floods of a rather dull yellow/brown pigment. Subsequent heating of the washed root to boiling point in a 10% solution of aluminium sulphate results in a true red colouration which can then be precipitated onto aluminium hydroxide to produce a pink/red lake pigment. The original procedure outlined by Rawson, Gardner and Laycock involved the use of lead acetate (79 parts /100 parts alum) applied to the red solution obtained after boiling in aluminium sulphate. This solution (now containing the lead acetate) is filtered leaving lead sulphate behind and a particularly bright crimson filtrate. I did, at a later stage follow this procedure in order to note any differences in the final precipitated lake. The inclusion of lead acetate in the process whilst not essential did seem to produce brighter cleaner colours, particularly after several stages of filtration.

## Indigo

In the literature review, I stated that of all the plant colours, Indigo, from whatever species it is derived, is completely unlike all other plant dyes. In very general terms, the colouring principle indigotin exists within the plant leaves as a colourless compound of indoxyl known as indican, the blue compound indigotin, forms only when the indican is exposed to oxygen which process also renders it insoluble in water. In fact, indigo has, to all intents and purposes, more in common with the insoluble pigments mentioned earlier than to any ordinary plant pigment. Whether it is derived from the Indigoferous, Polygonaceous or the Isatis species, the essential colouration and composition of the pigment is the same.

As a dry powdered pigment the colour of indigo is a dark blue and as a substitute for process blue, rather unsatisfactory. Fortunately there are two methods which can be used to achieve a brighter cleaner blue, one of which has the advantage of rendering the indigo pigment soluble in water.

## Sulphonated Indigo Extract

A variety of procedures for preparing a sulphonated extract of indigo are described in the literature each producing different degrees

of purity. The extract used in the print tests was prepared as follows:

1. 1 part powdered indigo was slowly added to a heat resistant container holding 5 parts concentrated sulphuric acid.
2. The mixture was stirred continually producing a smooth, lump-free solution, which was left to stand overnight.
3. Precipitated chalk was slowly added to the mixture, over a period of approximately 3 hours, until a pH value of around 6 was achieved.

The whole procedure was carried out in a fume cabinet, the extract being removed only when a relatively harmless pH value was attained.

The brighter, slightly redish-blue extract thus obtained is soluble in water and can be thickened in the normal way, or precipitated onto chalk, aluminium hydroxide etc. to form a kind of lake pigment.

A commercial product derived from synthetic indigo known as Indigo Carmine (CI 73015) and used in medical research, is produced in a similar way to sulphonated indigo. Samples of this product were

also prepared as thickened aqueous extracts and subjected to spectral reflectance and light fastness comparison tests.

### Maya Blue

According to the German pigment manufacturer *Kremer Pigmente*, the ancient Mayan people who inhabited the Yucatan Peninsula in modern Mexico employed hydrated silica (silicic acid) as a means of lightening indigo. The Mayas baked indigo with clay to achieve this lighter blue which is said to be more lightfast than the pure plant extract. The product obtained from Kremer, a form of Maya Blue, is certainly a lighter and brighter blue than unprocessed natural indigo as can be seen in appendix 5 though claims regarding it's improved lightfastness appear in these tests, not to be well founded.

The following chapter deals with the spectral reflectance and light fastness tests carried out on the samples described above.

## CHAPTER VIII

### Spectral Reflectance and Lightfastness Comparisons

The ability of the eye to recognise and distinguish between many thousands of different hues and tones is well known to anyone involved in Art and Design. Artists, designers and printers routinely make hundreds of fine colour distinctions in the course of a days work. Notwithstanding this highly developed skill, it is often necessary, for the purposes of objective analysis, to seek means of comparing colour samples which are not susceptible to the effects of mood, individual physiology or the particular environment in which the observations are made. In addition, the effect of what might be termed 'wishful thinking' or the researcher's unconscious desire to make favourable judgments in the light of a given hypothesis, cannot be ignored and should therefore be avoided where possible.

Spectral reflectance analysis provides one such objective means of determining exactly how similar or how different a particular pair of samples might be. It is also possible to accurately identify the particular spectral ingredients in a given sample and to identify possible modificatory measures which might bring the test sample into closer alignment with the standard.

Having prepared the printed samples referred to in chapter 3, I arranged to have the spectral analysis done by Dr Robert Withnall at

the University of Greenwich in the UK. These tests were restricted to the potential process colours, spectral analysis of black (being the absence of reflection) was considered unnecessary. It should be noted however that most blacks have some colouration within them, often leaning toward blue or red. Many of the samples were, even to the naked eye quite dissimilar to the process colour they were intended to mimic. Despite their obvious dissimilarity I thought it useful to have them analysed in order to ascertain their precise differences in the hope that the information gathered might help in any subsequent modificatory action.

Appendix 4 shows the spectral reflectance curves for a range of possible vegetable process colours along with the curve produced by a sample of the corresponding commercial process colour. Even a brief glance at these figures will demonstrate that none of the vegetable sources match precisely the process samples and indeed one might conclude from this that a four colour process print using vegetable sourced inks would be impossible to achieve. In the light of this observation it is interesting to note just how reprographically convincing some of the prints actually are (see Appendix 6).

The conclusion that may be drawn from this is that it is not necessary to have a true cyan, magenta, yellow and black in order to



reproduce a colour separation print with a reasonable degree of similarity to the original scanned artwork.

### Cyan

A comparison of the reflectance curve of natural indigo with a process standard (see p 6 and appendix 4)), demonstrates the fact that even with a considerable degree of dissimilarity, indigo is still capable of combining effectively with other vegetable process colours to reproduce a remarkably promising facsimile of the original (Fig.12).

The curves for the sulphonated samples are a distinct improvement over natural indigo and it may be that further improvement is not possible without resorting to some form of genetic modification of the plant species. Such a possibility is beyond the scope of this study, though others may find the suggestion intriguing enough to take up the challenge.

As has already been stated, the particular pigment used to produce cyan process inks is phthalocyanine blue, a slightly greenish blue derived from copper. The possibility of shifting a sulphonated indigo blue towards the green by adding yellow was considered and a series of admixtures were made using varying proportions of weld and persian berries.



Fig. 12. The original slide from which the artwork was scanned and separated (top) and the printed version of it (bottom). The plant sources used in this early example were not all lightfast and some fading has occurred.

Whilst a marginally increased similarity between the sulphonated indigo mixture containing approximately 10% weld or 5% persian berries and the process standard could be observed the resultant colour had a distinctly duller appearance. This is probably due to the presence of red in the indigo, which, when combined with yellow and blue would tend towards brown and thus create the dulling effect noted. Indeed this phenomenon is well known to artists who have long known that for maximum brilliance as few pigments as possible should be mixed together. A further complication would arise if the mixing of colours was adopted as a solution to the problem of creating a more faithful cyan. As well as cyan, the four colour process involves the use of yellow (along with magenta and black) to achieve its end and if yellow were also added to blue this would inevitably lead to an extremely complicated set of adjustments to the chromatic permutations of the four colours, leading to an even greater dulling effect. For this reason and in light of the relatively good results achieved with sulphonated indigo the spectral analysis of these mixtures was not considered useful. However, upon completion of the lightfastness comparison tests, certain factors became apparent which suggested the possibility of a reappraisal of the above observations and these will be discussed under light fastness.

## Magenta

The reflectance curves for the vegetable magenta samples provided rather more to choose from in terms of similarity to the standard process sample. Certainly Pokeweed, Brazilwood lake and the commercial preparation of rose madder are all within the magenta range, with pokeweed providing a near identical curve to process magenta up to the 600 nm point (see appendix 4, figs. MA - MD). We will see in the next section however, that pokeweed is particularly unstable in light and therefore not viable as a print ink of any permanence.

Though not a vegetable source, cochineal (MI) also compares well with the process magenta standard. In recent years, there has been a revival of interest in cochineal production as an industrial crop. Several cochineal farms surrounding the Mexican town of Oaxaca now produce significant quantities of cochineal dye where it is largely used in the production of carpets and rugs but also exported for use in cosmetics and food colourants. In addition at least one author (Rawson, Gardner and Laycock, 1916) notes that carminic acid, the principal colourant in cochineal, is also to be found in several plant species, citing *Monarda didyma* as an example. I obtained samples of the cochineal insect and a lake pigment prepared

from it (carmine) on a visit to the the *Centro Para Las Diffusion Del Conocimiento De La Grana Cochinilla*, in Mexico. As a related dye source, it seemed appropriate to demonstrate its potential in the context of this study.

Though the various madder lakes are less similar to process magenta than the sources previously mentioned, they do have the advantage of having a substantially greater resistance to the action of light. For this reason, of the sources looked at, a form of madder lake is considered the best alternative as a process magenta.

## Yellow

The spectral reflectance results obtained for the range of vegetable yellows suggest that a mixture of weld and persian berries produce the most faithful replication of standard process yellow. YC shows the reflectance curve for a 50/50 mixture of weld and persian berries and while the colour sample itself appears more saturated than the process standard, extensions of the ink are remarkably similar to the standard sample. Unfortunately, persian berries did not show sufficient stability in light to recommend their use in ink formulations (see lightfastness). This left weld as the sole vegetable alternative to

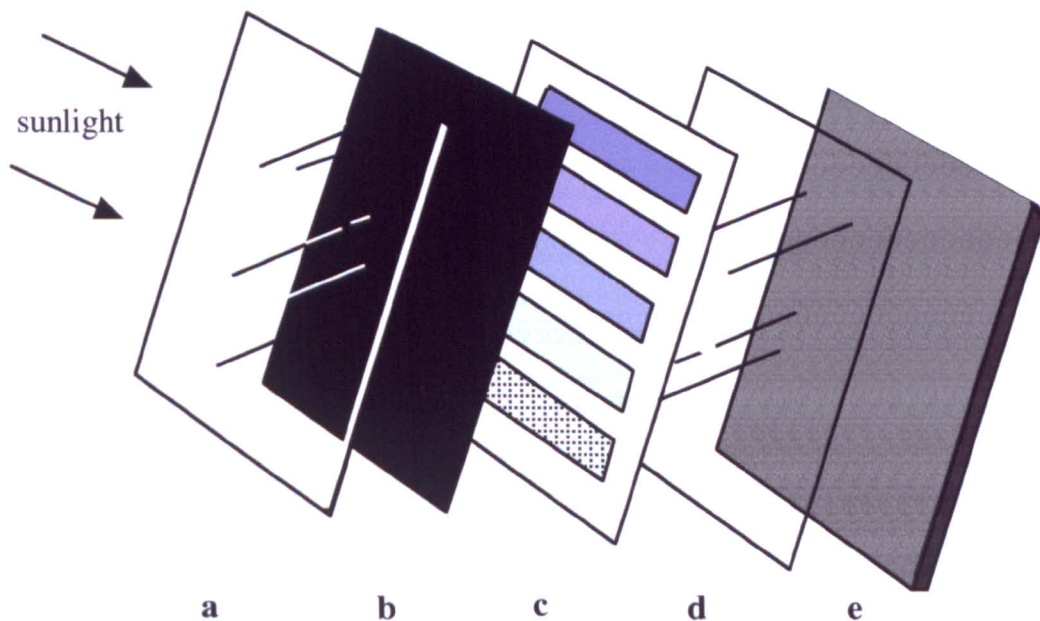
process yellow. Fortunately, the pure weld sample compares quite well with the standard process sample though there is considerably more reflectance in the blue/green area of the spectrum (400 to 480 nm) than would be ideal. Nevertheless, as with indigo, process yellow from weld is capable of reproducing reasonably faithful colour separation print results. Weld is said to be one of the most lightfast vegetable yellows known, so that in combination with madder and to a lesser extent sulphonated indigo (which in its sulphonated form is less lightfast than natural indigo) a reasonable compromise, in terms of spectral reflectance and lightfastness characteristics was arrived at.

### Lightfastness

In order to compare the behaviour in daylight of vegetable and standard process colours each sample was exposed to a total of 1000 hours of daylight graduated in 200 hour increments. The total amount of daylight over a period was calculated and divided into five equal parts using tables set out in Whitakers Almanac. The samples which were printed in strips measuring 6 x 1 inch with a mark indicating six equal divisions of the strip along the bottom edge. The print samples were placed on MDF board, the surface of which was protected by a sheet of acetate to prevent migration of acids from the board to the

samples. An occluding sheet of fully exposed PMT film (providing a high degree of opacity) was placed over the sample leaving one of the six divisions exposed. The samples were then covered with 2 mm picture glass attached with 'bulldog' clips. These boards were then placed in a south facing window at an angle of 52.5° to the horizontal so as to ensure optimum exposure throughout the period which extended from late January to mid April. After the expiry of the first 200 hour period the occluding sheet was moved laterally to expose the next division, and so on until the fifth division had received its allotted exposure. The final and sixth division was left unexposed so as to show the increasing effect of exposure from zero to the full 1000 hours. Fig. 13 illustrates the simple arrangement devised to ensure that all samples received equal amounts of exposure. In all, 11 potential cyans based on indigo, 8 yellows from weld and persian berries and 8 magentas from madder, brazilwood, pokeweed and cochineal were tested alongside three samples of standard process colours (Rowney System 3). Appendix 5 show the results of these tests

**Fig 13.** Simple arrangement for comparing lightfastness



- |          |                    |          |                          |
|----------|--------------------|----------|--------------------------|
| <b>a</b> | 2 mm picture glass | <b>d</b> | acetate protection layer |
| <b>b</b> | occluding sheet    | <b>e</b> | MDF backing board        |
| <b>c</b> | printed test card  |          |                          |



It should be stressed that these tests were not designed to ascertain a given sample's lightfastness per se, but rather to determine the comparative degree of fading or otherwise, with that of standard process samples under the same conditions. A comparative study of this sort has never been undertaken to the best of my knowledge, though there have been a number of studies to determine the lightfastness of vegetable colours on textiles and at least one on the fading of colourants used in Japanese block prints (see Literature Review). For this reason the use of highly sophisticated lightfast testing equipment of the kind used to determine near absolute lightfast values was considered inappropriate. It should also be noted that the tests described above subjected samples to full sunlight, conditions under which most fine art prints, for instance, would never be allowed to endure. It may be assumed that under average indoor lighting conditions all the samples tested would survive for considerably longer periods.

Some general observations regarding the lightfastness of the samples are made here, reference should also be made to the the full list of figures set out in appendix 5.

## Cyans

Tests CA through to CE (see appendix 5), all formulations of a sulphonated indigo lake prepared by the author, show very clearly that the pigment suspended in sodium alginate and gum arabic has far greater resistance to the action of light than those suspended in the commercial acrylic print medium. The reason for this is not easily understood. There may be some protective action exerted by the alginate/gum mixture which is not present in the acrylic print medium. There may equally be some form of chemical reaction taking place which has the effect of diminishing the indigo's resistance to light. One possible explanation, though far from satisfactory in my view, is connected with the comparative degree of transparency of the two media. The acrylic medium is virtually transparent, even in quite thick layers, the alginate/gum medium possesses some colouration even in thin print layers. This colouration may have the effect of screening the suspended pigment from the more destructive radiation in sunlight, though Padfield and Landi's study of so-called 'UV filtering' glass for use in museum conservation (admittedly under artificial light conditions) suggests that such filtering products have little practical protective effect on vegetable dyed samples (Padfield and Landi, 1966). The likelihood of my having stumbled upon an

effective UV filtering product in the shape of two historically well known and technically well understood materials seems remote. At any rate this remains one of the many so far unanswered questions posed by this study. The overall light fastness of sulphonated indigo lake when compared to the standard process cyan could be characterised as only moderately good, since over the same period the commercial colour exhibited far less fading. The commercially obtained indigo carmine prepared from synthetic indigo (which is nevertheless identical in structure to the purest forms of natural indigo) showed an encouraging degree of resistance to light. It is interesting to note that the samples containing a proportion of weld (CH to CK), after an initially marked degree of fading similar to that in the pure sample (CG), then remain noticeably more consistent throughout the remaining 800 hours of exposure. This may indicate that the presence of weld contributes to the photo-stability of sulphonated indigo. It has been stated, in the section on spectral reflectance, that these weld/persian berry/sulphonated indigo combinations, were not considered satisfactory, because of the dulling effect brought about through mixing. Whilst this remains true, the addition of small amounts of weld, perhaps even as low as 2 or 3% may have a similar stabilising effect, without producing significant

dulling of the resultant colour. Tests designed to verify this hypothesis have yet to be carried out.

### Magenta

Of the 8 magenta samples tested, only three displayed good resistance to fading in light. Brazilwood, both as a thickened aqueous extract and as a lake (MA and MG) both showed a very marked degree of fading even after the expiry of the first 200 hour exposure. Similarly, Pokeweed (MB and MC) and cochineal (MI) displayed extremely poor lightfastness. As was expected, the two commercial preparations of madder lake (MD and MH), along with the sample made by the author (ME) were all remarkably stable in light, though, as with the indigo/weld preparations, a distinct loss of colour appears after the first 200 hours, followed by near consistency over the remaining 800 hours of exposure.

### Yellow

Analysis of the photo-stability of the yellow samples is a little more complicated. Samples YF to YH, the weld and persian berry lakes precipitated on chalk, may be dispensed with quickly, since they each display a singularly poor resistance to light. Each of these

samples faded almost entirely within the first exposure period. It may be that better performance could be achieved by replacing chalk with aluminium hydroxide as the precipitate and this will be the subject of future work.

The behaviour of weld was a little disappointing given its reputation as the most lightfast yellow pigment known. The weld sample tested here (YB) shows a gentle but inexorable rate of fading over the whole exposure period. It may be possible to read too much into the fact that there is a distinct increase in the rate of fading at the 800 hour stage since variations of strong sunlight at any one time may be responsible. Nevertheless, the final exposure period seems to have diminished the colour markedly in all samples (YA to YE). As was stated in the section on reflectance, the best 'process' equivalent appeared to be YC (50% mix of weld and persian berries). In the lightfastness tests too, this sample seems to have survived best.

The persian berry sample (YA) shows a marked darkening effect upon exposure to light, quite the opposite to the expected fading, and this darkening appears also to be present in YC, but to a lesser degree.

It would seem that the 50/50 mix of persian berries and weld is the best all round source of vegetable process yellow, though the

distinct acceleration in fading after the 800 hour stage is far from ideal.

Lightfastness comparison tests have not so far been performed on the blacks derived from logwood and oak galls.

The conclusion arrived at after the reflectance and lightfast testing described above, i.e. that indigo, madder and weld/persian berry combinations are the best plant sources of process colour, holds, in many ways, few surprises. Constant reference to these sources in the literature suggests that they would indeed turn out to be the best candidates with regard to light fastness. The superiority of a weld/persian berry combination over straight weld is something of a surprise, given both weld's reputation for lightfastness and its greater 'naked eye' similarity to process yellow, particularly in lake form.

My early excitement over *Solanum guineense* (garden Huckleberry) as a source of cyan may, in the light of recent information, turns out to be more justified than I initially concluded. Following discussions with colleagues about the loss of my *Solanum* samples under refrigeration, I came to believe that continued testing of anthocyanin pigments may be something of a waste of effort. Every authority insisted upon the poor lightfastness of these pigments

so that I was becoming persuaded of the futility of continued investigation of these particular sources. However, Dr. Dominique Cardon of CNRS, Lasalle, France and Dr. Naceur Ayed of the Institut National des Sciences in Tunis, have both recently indicated to me the possibility of rendering the highly photo-unstable anthocyanin pigments extremely fast to light by the use of a technique known as co-pigmentation.

Unfortunately, this exciting new information comes too late to be included as part of this study and will have to form part of future work. If co-pigmentation techniques do significantly increase the lightfastness of anthocyanins, then not only will *Solanum guineense* become a possible alternative to indigo, but pokeweed too, a very important alternative to madder as a source of magenta.

## CHAPTER IX

### Health, Environmental, Economic and Related Issues



The impetus behind this study was simple curiosity - to determine the feasibility of producing CMYK screen printing inks from plant material. As the work progressed, several important factors became increasingly apparent; factors relating to the environment, health, economic and commercial practicality. These issues are discussed in this chapter

### The Environment and Occupational Health

Over the past 25 years, concern about the environmental damage caused by industry, particularly those industries which rely heavily upon oil and coal, has become a major source of public concern in the industrialised countries of the west. The literature, both popular and academic, abounds with studies and reports demonstrating the extent to which industry has paid scant attention to the effects of its activities on the environment, the health of its own employees and the public at large. The United Nations Conference on the Environment held in Rio de Janeiro in 1992 and attended by heads of government from around the world, made several environmental policy commitments that represented a sea-change in official opinion. For the first time, the health of the planet and its inhabitants were placed high on the political agenda. Environmentalism, a term that a

little over 25 years ago had all kinds of crankish connotations in the minds of many, had now become a matter of serious widespread public concern. Initiatives like those that prompted government backed schemes for producing electrical power from wind and wave energy, the increase in energy conserving appliances, the spread of recycling programmes and the attempts at reducing the use of non-renewable sources of energy (oil and coal), continue to be made at every conceivable technological level.

These efforts are not necessarily restricted to the sophisticated industrialised nations. Small rural communities in Bangladesh, Haiti or Zimbabwe are just as likely to be utilising some form of renewable technology as any 'right thinking' inhabitants of the U.S.A., Denmark or Sweden. *Appropriate Technology*, the journal of Intermediate Technology, the international sustainable development organisation, has spent the last 25 years promoting the low-tech/low energy approach which exploits local technologies and natural resources and this is often the lowest cost option. For many countries burdened with international debt through importation of western 'resource intensive' technologies<sup>1</sup>. Similar organisations and activities around the world

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<sup>1</sup> *Appropriate Technology* along with *Waterlines* and *Small Enterprise Development* is as well as about 30 new books each year on all aspects of development and appropriate technology has been an invaluable source of reference for practical sustainable initiatives since its foundation in 1966 by Dr E.F. Schumacher, author of *Small is Beautiful*. It is published by the Intermediate Technology Development Group 103-105 Southampton Row London WC1B 4HH, UK.

demonstrate clearly that the realisation that our everyday existence depends entirely upon the use of raw materials which will eventually become prohibitively expensive through scarcity, is no longer one reached only by a few academics. Ordinary people the world over now appreciate the need to think afresh about the resources we use to sustain our modern way of life.

Problems relating to the environment and public health are inextricably linked and a discussion of one inevitably leads to discussion of the other. Indeed it is often not possible to determine which is responsible for improvements in industrial practice when they occur - cynics would say that the real concern is fear of litigation - whatever the reasons, manufacturers have begun increasingly to turn their attention to health, safety and the environment over recent years.

For their part, ink and paint manufacturers around the world have sought to develop water-based products that do not require the use of hazardous and non-renewable petroleum based solvents for thinning and cleaning or which use high proportions of petroleum based oils as pigment media. The screen printing industry has been among the leaders in this 'water-based revolution' and there have been undoubted benefits resulting from their initiatives. Life in the

‘enlightened’ screen print workshop has improved immeasurably. Toxic and noxious fumes from solvent based inks, along with the often serious medical conditions associated with them, are slowly being reduced. A number of researchers around the world have worked to bring the dangers of traditional techniques to the attention of those involved particularly in fine art printmaking, where techniques dating back 500 years are still being used despite their often fatal consequences.

Keith Howard, founder of the Canadian School for Non-toxic Printmaking in Alberta and now Rochester Institute of Technology (NY) cites the astonishing conclusion of the Alberta Labour, Occupational Health and Safety Division report of 1978 as follows: ‘...there are 112 toxic and noxious substances commonly used by the practitioners of traditional printmaking media. Some of these chemicals and solvents are: nitric, hydrochloric, sulphuric, hydrofluoric and chromic acids; toluene, benzene, turpentine, lacquer thinner, methyl cello solve acetate, xylene and aromatic hydrocarbon solvents. Some of the ‘known’ occupational side effects from exposure to these chemicals, many of which are carcinogens, are; birth defects, central nervous system damage, benign pneumoconiosis, asthma, emphysema, severe burns from which gangrene may follow,

systemic poisoning which affects the lungs, liver, kidneys, heart, nervous disorders, skin, eruptions, dermatitis and damage to the mucous membranes and upper respiratory tract. Because most traditional printmaking studios use many of these products simultaneously, the accumulated effect and the resulting health damage related to combining these toxins is unknown” (Howard, 1997).

Workers like Howard, the originator of a whole range of non-toxic printmaking techniques, his assistant Elizabeth Dove, Friedhard Kiekeben, inventor of *Edinburgh Etch*, an acid free etchant, George Roberts of the University of Boise Idaho and his non-toxic polyester plate lithography, Henrik Bøegh, Danish printmaker and disseminator of the non-toxic ‘credo’, have all demonstrated not only that printmaking can be a far healthier activity than it has traditionally been but that it can also be a considerably cheaper one. Henrik Bøegh cites figures produced by Edinburgh Printmakers Workshop in 1995 demonstrating an overall saving on all consumable materials of 62% over a period of 2 years (Bøegh, 1998). This kind of evidence along with the obvious reduction in toxic material use has contributed to a growing demand from practitioners around the world for a cleaner more eco-friendly approach to the art form.

In my own field, screen printing, the introduction of ‘water-based’ inks at the beginning of the 1990’s at first provoked a mixed reception from practitioners. To some, like Steve Hoskins of the University of the West Of England, it presented an exciting challenge, to others, wary of new methods and materials (particularly ones that might render cherished techniques redundant), it presented a threat. Although I joined the chorus of approval when the new water-based acrylic screen printing inks were first introduced, I was not entirely convinced by the so-called ‘water-based’ revolution. The use of these new ink formulations did mean that for the first time screen printers did not have to use any of the hydrocarbon and other volatile solvents responsible for so much damage to health, but these new inks still relied on acrylic copolymer technology for viscosity, and curing.

Acrylic copolymers are produced from fossilised hydrocarbons - coal and oil. Stephens (1996) lists what he calls a ‘typical formulation’ for commercial ‘water-based’ screen inks as follows:

Pigment	15%
Carboxylated acrylic emulsion	45%
Styren/acrylic emulsion	15%
Ammonia	0.5%
Microcrystalline wax emulsion	5%
Silicone defoamer	0.5%

Levelling agent	1%
Dowanol DPM	4%
Water	14%

The above formulation demonstrates the unavoidable fact that in excess of 60% of the ‘typical formulation’ of a water-based screen ink is in fact wholly or partly derived from petrochemical products. When one considers that these same raw materials are also the source of many of today's synthetic pigments, one is forced to conclude that problems relating to the environment, sustainability and public health, remain fundamentally unanswered.

### Historical Precedent and The Economics of Sustainability

The possibility of reintroducing a technology that uses ‘un-fossilised’ hydrocarbon material (ie. living plants) for the production of pigments and thereby rendering the product more genuinely water-based and sustainable, may seem at first sight to be a retrogressive step.

Prior to the mid-19th century, when synthetic colours began to be developed in increasing numbers, plants were one of the chief sources of used in the textile dyeing and printing industry. Indeed colours from plants of one sort were in use throughout history and across cultural and geographical divides. From Scythian burial

mounds on the borders of South-West Mongolia, textiles preserved in ice and dating from the fifth century BC reveal an extensive and sophisticated use of vegetable dyes (Eiland,1979). Saffron (*Crocus sativus*), well known as the source of a truly brilliant if rather fugitive yellow dye, has been shown to have been used both as a colourant and medicine, at the time of the the Greek and Persian civilisations (J. & M. Cannon,1994). Pliny, in his *Natural History* speaks of vegetable dyeing in Egypt during Roman times and it is thought that these people could have learned their craft from India, where textile dyeing had reached a position of pre-eminence. Indian skill in vegetable dyeing and painting reached a high point in the two centuries from 1600 to 1800 A.D., when the painting and resist dyeing of cotton cloth, known to us as Chintz, became the basis of the largest trade in textiles that the world had ever seen (Irwin & Brett, 1970). In mediaeval and renaissance Europe treatises and manuals on the preparation of artists colours contain many references to plant colours, one such being Cennini's, 'Il Libro dell' Arte' of 1437, in which he sets out recipes for the preparation of block-printing inks from saffron and brazilwood (*Caesalpinia sp.*). The Strasbourg manuscript, of an earlier period, also describes the use of a whole range of plants used in the manufacture of inks and watercolours.



There were developments in vegetable block-printing inks in 17th and 18th century Japan, (though the Japanese use of vegetable colours in dyeing goes back much earlier than this), where it is interesting to note that some colours were actually leached from previously dyed (indigo) cloth (Strange, 1924). Throughout mediaeval Europe there was extensive cultivation of woad (*Isatis tinctoria*), the well known source of a blue dye said by Caesar to have been used by the ancient Britons to colour their bodies, though whether this practice was medicinal or warlike in intention is open to dispute (Goodwin, 1982). Even today, South American Indians use plants like Achoite or Annato (*Bixa orellana*) for much the same purpose (Uphof, 1959) along with a range of other native species like Muicle o mohuitli (*Jacobinia spicifera*) and the insect and animal dyes obtained from the cochineal scale insect (*Dactylopius coccus*) and the marine molluscs *Purpura pansa* and *Murex* (Velasco Rodriguez, 1995). The beautiful coloured rugs and blankets produced by the Navaho (though historically speaking, these examples are a relatively recent art form), form just a part of a long tradition of plant colour use by the native inhabitants of North America. Indigenous species like bloodroot (*Sanguinia canadensis*), prickly pear (*Opuntia sp.*) and golden seal (*Hydrastis canadensis*) were used to dye and stain cloth, skin and

basketry (Krochmal, 1974).

Clearly, there is a huge historical precedent for considering vegetable colours as a serious alternative to present day sources, and their production on an industrial scale is by no means beyond the bounds of possibility. In 1864, upwards of 40,000 tons of logwood (*Haematoxylon campechianum*) was imported into the United Kingdom. Around 19,000 tons of madder (*Rubia tinctoria*), 13,000 tons of sumac (*Rhus coriaria*) and 3,000 tons each of indigo (*Indigofera tinctoria*) and Quercitron oak bark (*Quercus velutina*) were also imported into Britain during the same period (Rhind, 1872). The same kinds of figures for these, and a whole range of other dye-plants, can be assumed for the textile industries of other industrialised countries at that time.

It can be readily seen then, that vegetable sourced colours were commercially important in the pre-synthetic era. And yet within the space of around forty years from the discovery of a mauve 'aniline' dye derived from coal-tar by W.H.Perkin in 1856 (Ponting, 1981), the vegetable colour industry was virtually dead. Fortunately the dyers and chemists of the period were painstakingly meticulous in their recording of detail, and a whole host of books were published

throughout the 18th and 19th centuries outlining the procedures involved in vegetable colour extraction. Principal among these was the magnificently titled, *Experimental Researches Concerning the Philosophy of Permanent Colour* by Edward Bancroft, published in 1813. In the U.S. too, early dyers were publishing details of their methods in order to improve local industry, and reduce reliance on old world imports and technologies. One such U.S. pioneer, Elijah Bemiss published *The Dyers Companion* in 1806, and this work continues to be available to this day.

The concentration of colour in a given quantity of coaltar or oil is many times that of an equivalent quantity of plant material. In addition, the acreage needed to cultivate industrial quantities of colour producing plants would far out-measure the surface area currently taken up by the oil and coal industries. On the other hand, when one considers that vast tracts of good agricultural land in Europe and the U.S. currently lay fallow year after year as a result of government policies relating to over production, there would appear to be no reason why we could not utilise this land, along with the energies of otherwise unemployed agricultural workers, for the production of

dye-plants<sup>2</sup> . Indeed, there are now enough encouraging indications from government and industry to make the proposition worthy of serious consideration.

Several initiatives currently being pursued throughout Europe and the U.S. demonstrate the seriousness with which governmental institutions have taken the issue of vegetable sourced alternatives in the dyeing and printing industries. In a recent article by Dr. Dominique Cardon (Cardon,1996) on the historical use of natural dyes, she points to a European Community funded Anglo-Italian project to investigate the agricultural and processing possibilities of natural dye plants including woad, weld and madder. Two British researchers, David Cooke and Kerry Stoker, have also recently obtained substantial funding from the U.K. Ministry of Agriculture Fisheries and Food, to investigate the use of woad in the production of ink jet printing inks. A farm-scale pilot project is currently underway the aim of which is to determine the viability of agricultural production of woad, which, in combination with Cooke and Stokers 'continuous extraction' technique has attracted £700,000

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<sup>2</sup> Under the UK Ministry of Agriculture Fisheries and Food Arable land Payments scheme some 295,000 hectares of arable land was 'set aside' for non-production in the UK in 1997. Compensatory payments of between £214 and £306 per hectare were thus made to farmers in order to grow nothing! (MAFF [http://www.maff.gov.uk/farm/schemes/aap\\_sa.htm](http://www.maff.gov.uk/farm/schemes/aap_sa.htm))

worth of government and industrial development funds.<sup>3</sup>

In the field of pigment vehicle technology, research commissioned by The American Newspaper Association as early as 1985, resulted in the formulation of what became known as ANPA-INK, a product in which 'tall oil' (derived from wood pulp) was substituted for the traditional petroleum based media.<sup>4</sup>

The United States House of Representatives Vegetable Printing Act, of 1994, aims to replace, at least a proportion of the petroleum-based oils in lithographic inks used on government documents, with a vegetable oil. The increased use of vegetable oils would, 'reduce reliance upon non-renewable (petroleum) energy resources; use fewer environmentally damaging products; reduce volatile organic compound emissions; and increase the use of renewable agricultural products'. The act goes on to say that a domestic demand for 2,500M lbs of vegetable crop, producing 500M lbs of oil, would be created. One possible source of all this vegetable oil, based on research commissioned by the Newspaper Association of America, or NAA), is the massive surplus of soya beans grown in the U.S. - 1.106 million metric tons in 1992<sup>5</sup> .

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<sup>3</sup> Reported in the Financial Times September 16th 1999.

<sup>4</sup> Reported in The Ink Company's *Soybean Oil Technology and News Inks*  
<http://www.ink.net/soynews.html>

<sup>5</sup> Reported in The Ink Company's *Soybean Oil Technology and News Inks*  
<http://www.ink.net/soynews.html>

Clearly, there is no suggestion here that the cultivation of vast quantities of vegetable material would be impractical.

How much more beneficial would it be, if the actual pigmentation of these inks could be derived from 'renewable agricultural products' like indigo, madder and weld?

In Europe, the German paint manufacturer *Auro*, has been manufacturing a range of vegetable paints for some time at a reasonable cost. Though their production levels are only modest when compared to the giants of the paint industry, their highly sophisticated production plant does demonstrate that these raw materials can be subjected to modern industrial scale production methods (*Auro Pflanzenchemie GmbH, Braunschweig Germany*). The step from paint to ink is a short one, so that the commercial possibilities of producing these colours on a larger scale would seem to be more a question of will, rather than of practicability.

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## CHAPTER X

### Concluding Summary

In the introductory chapter, I identified several questions that would be addressed during the course of my investigation. What colours might be obtained from plant material? How suitable might these colours be within the context of the study aims? How might they compare to existing ink pigments and what if any advantages might be derived from their use? The following chapter is a summary of the my conclusions to those questions.

### Sources of Colour

Using the considerable body of literature on textile dyeing as well as extensive testing of potential plant sources, it was possible to identify several sources for each of the four process colours. A workable cyan substitute could be extracted from indigo or woad and this 'natural' form could be improved using sulphonation or the process resulting in so-called 'Maya Blue'. The best source of yellow was found to be a mixture of weld and persian berries. A lake pigment derived from madder root, though more difficult to prepare, proved to be the best practical substitute for process magenta. Black from logwood provided perfectly satisfactory results when printed. Several other possible sources were discounted chiefly due to their instability in light or varying pH conditions. With the exception of

logwood and indigo, these sources are all viable as crops in the UK and Europe and were all grown in the Cat Hill Ink Garden at Middlesex University.

### Chromatic Comparison with Existing Process Colours

Spectral reflectance comparison tests were made to determine the similarity of sources which seemed to the naked eye to present possible alternatives to the commercial range of process colours. The sources outlined above were seen to resemble most closely the commercial range, though none could be said to be an exact match. The persian berry/weld combination proved most similar to its process yellow equivalent and whilst pokeweed also compared well with process magenta it was found to be deficient in terms of light fastness. Indigo from any source (*Indigofera* spp., woad etc) was least chromatically similar to process cyan. In sulphonated form, or in the form known as Maya blue, a distinct improvement in reproductive quality was noted.

### Light fastness

While all sources did not perform as well as any of the standard process colours when exposed to daylight, they nevertheless displayed

a surprising degree of resilience considering the generally accepted view that vegetable colour has poor light fastness. After 1000 hours of south facing exposure both unprocessed indigo and Maya blue compared most favourably with its process equivalent. The persian berry/weld and madder lake preparations displayed a lesser degree of light fastness though still within acceptable limits. Certainly, for print applications that do not require the very highest degree of resistance to light, such as short life commercial work, packaging, indoor poster work, books and brochures, they present a practical alternative to existing commercial inks.

### Ink Formulation

A practical screen printing ink, should have a consistent degree of viscosity, it should also be capable of adhering to the substrate without forming a discontinuous or unstable layer when dry. The combination of sodium alginate and gum arabic provided the best formulation for an ink which satisfied the criteria necessary for a usable water-based screen printing ink. The flow characteristics, viscosity and drying time of the vegetable inks compared favourably with their commercial equivalents. The addition of anti-bacterial and antifungal agents were found to increase shelf life of the product

considerably - samples survived uncontaminated for periods in excess of 18 months as compared with untreated samples which deteriorated markedly after a matter of weeks.

### Advantages and Disadvantages

The paint and ink industry relies heavily upon entirely non-renewable raw materials in the production of both colour and media. The essentially renewable nature of plant based pigments and media in a world which is increasingly concerned about the use of finite natural resources is an obvious advantage over the existing non-renewable sources of colour. The introduction of agricultural scale production of colour producing plants has been shown by other researchers to be viable and government agencies around the world have expressed keen interest in encouraging this kind of use, particularly where land previously 'set-aside' for non-production may be utilised.

The health and safety of workers in both the manufacture and use of plant based inks is demonstrably improved. Toxic and noxious substances such as toluene, benzene, turpentine, lacquer thinner, methyl cello solve acetate, xylene and aromatic hydrocarbon solvents, are all absent from the new process.

Entirely waterproof formulations however, were not found to be possible without the introduction of acrylic polymer or vegetable oil media. Light fastness too, remains a problem where the very highest standards of photo-stability are required.

The visual component of this study, a series of 35 four-colour reproductions of plant sources and related material along with the test prints reproduced in appendix VI, provides a practical demonstration of the effectiveness of plant based inks. Several problems remain however, the search for a waterproof or water-resistant formulation which does not rely upon non-renewable raw materials, being the most important area for further development. In addition, further work on light fastness, including an investigation of some of the latest commercial additives said to improve photo-stability will be undertaken.

\* \* \*

Throughout the period of this study I have published several articles, taken part in radio and television broadcasts, given lectures and presentations to conferences, conducted workshops and held

exhibitions connected with my work. These activities are listed below.

### Publications

- 1995 'Fruit and Veg or Growing Your Own Ink' *Printmaking Today* Vol.2,4.
- 1995 'Fruit and Veg' *The Point* Vol.1 No.1
- 1995 'Grow Your Own Inks' *Appropriate Technology* Vol.22 No.2 1996
- 1995 'Grow Your Own Inks' *Screen Process* Vol.45 No.9
- 1997 'Adventures in the Ink Garden' *Museum of Garden History Newsletter*.
- 1997 'Phytochromography - Printing with Plants' *Printmaking Today* Vol 6 No 3
- 1999 'Phytochromography' *Leonardo* Vol 32, No. 1, MIT Press, USA

### Broadcasts

- 1995 BBC TV *Tracks* (BBC2) 'A Brush With Nature' Interview and demonstration.
- 1995 BBC World Service. 'Future perfect' extended interview on dye plants and their applications in print for broadcast to the countries of the Pacific rim.

### Lectures

- 1996 *Development of Vegetable Sourced Inks for Use in Screen Process Printing Inks* presentation to the Annual Conference of Dyes in History and Archaeology 15, Manchester University, UK.
- 1996 *Phytochromography* presentation to the Research Network in Art and design Conference, 'Evolution in Art and Design' Middlesex University, UK.
- 1997 *New Developments in Four Colour Printing using Plant Colourants* presentation to the Annual Conference of Dyes in History and Archaeology 16, Musée Historique des Tissus, Lyon, France.
- 1997 *Growing Your Own Ink* Conference/Seminar on Non-toxic Printmaking Helsingør, Denmark.
- 1998 *Spectral Reflectance Analysis of Plant-based CMYK Printing Ink Substitutes* jointly presented with Dr Rob Withnall to the Annual



1998 *Spectral Reflectance Analysis of Plant-based CMYK Printing Ink Substitutes* jointly presented with Dr Rob Withnall to the Annual Conference of Dyes in History and Archaeology 17, National Maritime Museum, Greenwich, London.

1999 *Phytochromography - Adventures in the Ink Garden*. 'Linneveckan i Uppsala' (Linnaeus Week in Uppsala) The Gustavianum, University of Uppsala, Sweden.

### Workshops

1997/8 Canadian School for Non-toxic Printmaking, Grande Prairie, Canada.

1999 March and August, Grafikens Hus, Centre for Printmaking, Mariefred, Sweden.

### Exhibitions

1997 *Adventures in the Ink Garden*. Museum of Garden History, London, UK.

1997/8 Pro Arts 1997 East Bay Open Studio Show Oakland California USA.

1997 The Printmakers Council Exhibition, Whitely's Gallery, London, UK.

An exhibition of my own prints along with those of my students at Grafikens Hus, Sweden, is currently under preparation and is scheduled for 27 May - 17 September 2000.

Reference to my work is also cited in the following publications:

1997 Prints published in *Journal of the Society of Dyers and Colourist* Vol 113, April 1997 pp 117 and 120 .

1997 Prints published in article by Dr.Dominique Cardon in *Polyphenols 96* , Vol 6 No 3 Autumn 1998, France.

1999 Mackwitz, Hans-Werner, 1999. *Kompendium Nachwachsener Rohstoffe* (Compendium of natural resources) Ecomed, Landsberg, Germany.

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Middlesex - <http://www.mdx.ac.uk/www/vcd/21PS/PSphyto.html>

Beth Lacour Yavapai College • 1100 East Sheldon • Prescott AZ-

[http://www.yavapai.cc.az.us/division/v&p\\_arts/art/lacour/Veg-Ink/VegInk.html](http://www.yavapai.cc.az.us/division/v&p_arts/art/lacour/Veg-Ink/VegInk.html)

Kala Institute - <http://www.kala.org/phytochromography.html>

## **Appendix 1 Plant Sources Described in the Literature**

The plants listed below have been categorised under three general headings, yellows, orange/reds and violet/blues. In practice, many provide hues which have a distinct tertiary element within them, yellows for instance often lean towards ochre, likewise reds are often better described as rust, and are thus unsuitable for consideration as possible process colours.

## Yellows

*Aegle marmelos* f. Rutaceae (Bengal Quince/Wood apple)

*Agrimonia eupatoria* f. Rosaceae

*Alkanner tinctoria* f. Boraginaceae (Alkanet)

*Allium cepa* f. Liliaceae (Onion)

*Amygdalus communis* *A. amarus* f. Rosaceae

*Anogeissus leiocarpus* f.

*Anthemis tinctoria* f. Compositae (Camomile)

*Artemisia vulgaris* f. Compositae

*Asperula tinctoria* f. Rubiaceae

*Aster amellus* f. Compositae (Star wort)

*Berberis vulgaris* f. Berberidaceae (Barberry)

*Bidens tripartita* f. Compositae (Water Agrimony)

*Buddleja davidii* f. Loganiaceae (Buddleia)

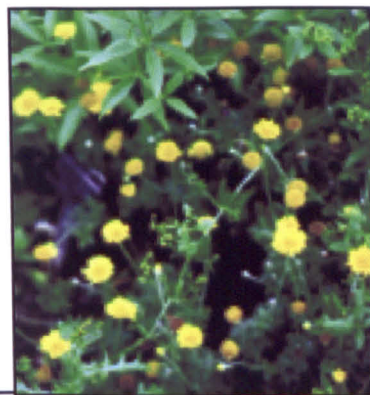
*Carthamus tinct.* f. Compositae (Safflower)

*Ceanothus americanus* f. Rhamnaceae

*Cheiranthus cheiri* f. Crucifereae (Wallflower)

*Chlorophora tinct.* f. Moraceae (Fustic)

*Citrus aurantifolia* f. Rutaceae (Lime)



Citrus limona f. Rutaceae (Lemon)

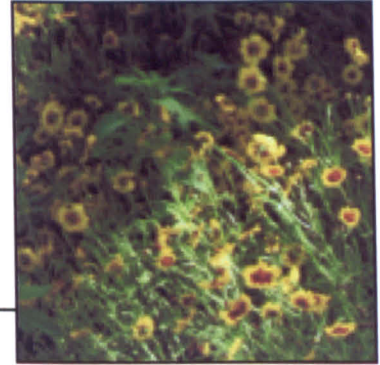
Cochlospermum tinct./planchoni/i f.Cochlospernameae

Combretum glutinosum f. Combretaceae

Convallaria majalis f. Liliaceae (Lily of the Valley)

Convolvulus soldanalla f. Convolvulaceae (Bindweed)

Coreopsis tinctoria and sp. f. Compositae



Cotinus coggygia f. Anacardiaceae (Smoke Tree)

Crocus sativus f. Iridaceae (Saffron)

Cucumis melo f. Cucurbitaceae (Melon Seeds)

Curcuma longa f. Zingiberaceae (Turmeric)

Daphne augustifolia, D. mezereum f. Thymelaeaceae

Delphinium zalil f. Ranunculaceae (Larkspur)

Ephedra pachyclada f. Ephedraceae (Ephedra)

Euphorbia sp. f. Euphorbiaceae (Milkwood or Spurge)

Fraxinus excelsior f.Oleaceae (Ash)

Garcinia hanburyi f. Guttiferaceae (Gamboge)

Genista tinctoria f. Leguminosae (Dyers Greenwood)



Glycyrrhiza glabra f. Leguminosae (Liquorice)

Helianthus annus f. Compositae (Sunflower)

Hydrastis canadensis f. Ranunculaceae (Golden Seal)

*Hypericum perforatum* f. Hypericaceae (St Johns Wort)

*Lawsonia alba*, syn. *L.inermis* f. Lythraceae (Henna)

*Ligustrum ovalifolium*, *L. vulgare* f. Oleaceae (Privet)

*Morinda citrifolium* f. Rubiaceae (Brimstone Tree)

*Pistacia terebinthus*, *P. chinensis* f. Anacardiaceae (Pistacio Tree)

*Polygonum persica* f. Polygonaceae (Arsemarkt)

*Prunus armeniaca* f. Rosaceae (Apricot leaves)

*Pteridium aquilinum* f. Polypodiaceae (Bracken)

*Punica Granatum* f. Punicaceae (Pomegranite Rind)

*Quercus velutina* f. Fagaceae (Oak Bark)

*Reseda luteola* f. Resedaceae (Weld)

*Rhamnus saxatilis* & spp. f. Rhamnasceae (Buckthorn)

*Rheum palmatum* / *R. rhaponticum* f. Polygonaceae (Rhubarb)

*Rhus cotinus*, *R. succedanium*, *R. glabra* f. Anacardiaceae (Sumach)

*Salix* sp. f. Salicaceae (Willow leaves)

*Sanvitalia* sp. Compositae (Zinnia)

*Serratula tinctoria* f. Compositae (Dyers Savoury / Saw Wort)

*Solidago virgaura* and *canadensis* f. Compositae (Golden Rod)

*Sophora japonica* f. Leguminosae (Japanese Pagoda Tree)

*Symphoricarpus rivularis* f. Caprifoliaceae (Snowberry)



*Tagetes* spp. see also 'Calendula' f. Compositae (Marigold)

*Tamarindus indica* f. Leguminosae (Tamarind Leaves)

*Tanacetum vulgare* f. Compositae (Tansy)

*Trionella foenum-graecum* f. Leguminosaceae (Fenugreek)

*Urtica dioica* f. Urticaceae (Nettle)

*Verbascum thapsus* f. Scrophulariaceae (Mullein)

*Viola odorata* f. Violaceae (SweetPansy)

*Vitis vinifera* f. Vitaceae (Vine Leaves )

## **Orange /Red**

*Aesculus pavia* f. Hippocastanaceae (Red Buckeye)

*Allium cepa* f. Liliaceae (Red Onion)

*Alkanna tinctoria* f. Boraginaceae (Alkanet)

*Alnus fruticosa* and sp. f. Betulaceae (Alder)

*Arctostaphylos uva-ursi* f. Ericaceae (Bearberry)

*Asperula tinctoria* (Dyers Woodruff)

*Beta vulgaris* f. Chenopodiaceae (Beetroot) —————

*Bixa orellana* f. Bixaceae (Annatto or Achiote)

*Brassica oleracea* f. Crucifereae (Red Cabbage)





Caesalpinia echinata, C. crista, f. Leguminosae (Brazil Wood)

Carthamus tinctoria f. Compositae (Safflower) \_\_\_\_\_

Chlorophora tinctoria f. Moraceae (Fustic)

Chrozophora tinctoria, syn Croton tinc. (Turnsole)

Codiaeum f. Euphorbiaceae (Croton Draco)

Crataegus canadensis, C. oxyacantha f. Rosaceae (Hawthorn)

Cruciata (laevipes?) f. Rubiaceae.

Galium spp. f. Rubiaceae (Bedstraw) \_\_\_\_\_

Geranium sylvaticum f. Geraniaceae (Wood Cranesbill).

Glaucium flavium/luteum f. Papaveraceae (Horned Poppy)

Haematylon campechianum f. Leguminosae (Logwood)

Hibiscus sabdariffa f. Malvaceae (Jamaica Sorrel)

Hypericum perforatum f. Guttiferae (St Johns Wort) \_\_\_\_\_

Juniperus communis f. Cupressaceae (Juniper)

Ligustrum ovalifolium, vulgare f. Oleaceae (Privet)

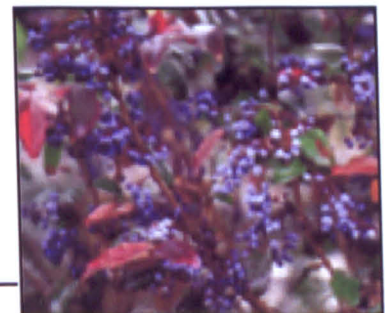
Lithospermum hispidissima syn. Arnebia h. f. Boraginaceae

Mahonia aquafolium f. Berberidoceae (Mahonia / Oregon Grape) \_\_\_\_\_

Morinda citrifolium/ trifolium /bracteata f. Rubiaceae (Brimstone Tree)

Morus nigra (Black Mullberry) f. Moraceae (Brimstone Tree)

Opuntia ficus-indica / O. Imbricata f. Cactaceae (Prickly Pear) \_\_\_\_\_





Parthenocissus quinquefolia f. Vitaceae (Virginia Creeper)

Phytolacca americana, P. decandra f. Phytolaccaceae (Pokeweed) —

Potentilla anserina f. Rosaceae (Cinquefoil)

Prunus laurocerasus f. Rosaceae (Cherry Laurel)

Prunus lusitanica f. Rosaceae (Portugese Laurel)

Pterocarpus santalinus f. Leguminosae (Red Saunders)

Rhamnus perseus f. Rhamnaceae (Buckthorn)

Rheum Palmatum f. Polygonaceae . (Rhubarb)

Rubia tinctoria, R. peregrina, R. cordifolia f. Rubiaceae (Madder) —

Rubus fruticosus f. Rosaceae.(Blackberry)

Sambucus nigra f. Caprifoliaceae (Elder) \_\_\_\_\_

Sanguinaria canadensis f. Papavaraceae (Bloodroot/Puccoon)

Sorghum caudatum f. Graminaceae (Guinea Corn)

Spartium scoparium f. Leguminosae (Common Broom)

Thespesia populnea f. Malvaceae (Tulip Tree/Portia Tree)

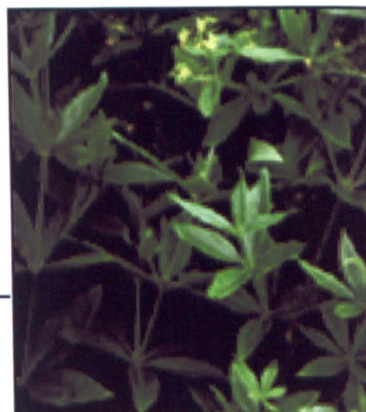
Vaccinium corymbosum f. Ericaceae (Blueberry)

Vaccinium macrocarpum f. Ericaceae (Cranberry)

Viburnum opulus f. Caprifoliaceae (Cranberry Tree)

Zizyphus nummularia/xylocarpus f. Rhamnaceae (Jujube)

Zonal Pelargonium f. Geraniaceae



**Violet / Blue**

*Amorpha fruticosa* f. Leguminosae (False Indigo)

*Baptisia tinctoria*/*B. australis* f. Leguminosae (False Indigo)

*Centaurea cyanus* f. Compositae (Cornflower)

*Cheiranthus cheiri* (syn. *Erysimum Cheiri*) f. Crucifereae (Wallflower)

*Chrozophora tinctoria*, syn *Croton tinc.* (Turnsole) f. Euphorbiacia

*Commelina communis* f. Commelinaceae

*Condalia obovata* f. Rhamnaceae (Bluewood)

*Cornutia pyramidata* f. Verbenaceae

*Cybistax sprucei* f. Bignoniaceae

*Datura fastuosa* (syn. *D. alba*) f. Solanaceae

*Dianella lavarnum/nemorosa* f. Liliaceae

*Elaeocarpus hookerianus/dentatus* f. Eleocarpaceae

*Eupatorium indigofera* f. Compositae

*Fraxinus excelsior/quadrangulata* f. Oleaceae (Ash)

*Fuschia macrostemma* f. Onagraceae

*Garcinia livingstonii* f. Gutterifaceae

*Geranium sylvaticum* f. Geraniaceae (Wood Cranesbill)

*Haemotoxylon campechianum* f. Leguminosae (Logwood)



*Helianthus annuus* f. Compositae (Sunflower)

*Indigofera tinctoria*/anil/arrecta Leguminosae (Indigo)

*Innula helenium* f. Compositae (Elecampane)

*Iris pseudacorus* f. Iridaceae (Yellow Flag)

*Isatis tinctoria* f. Cruciferaeae

*Jacobinia spicigera* (syn. *Justica spicigera*) f. Acanthaceae

*Lithospermum erythrorhizon* f. Boraginaceae

*Lonchocarpus cyanescens* f. Leguminosae

*Mahonia aquifolium* f. Berberidaceae (Mahonia/Oregon Grape)

*Marsdenia tinc.* f. Asclepiadaceae

*Mercurialis annua*, *M. leiocarpa* f. Euphorbiaceae

*Nerium tinctorium*/oleander f. Apocynaceae

*Polygala javana* (syn. *tinc.*) f. Polygalaceae

*Polygonum tinctorium* f. Polygonaceae (Japanese knotweed)

*Prunus domestica* f. Rosaceae (Plum)

*Prunus laurocerasus* f. Rosaceae (Cherry Laurel)

*Prunus spinoza* f. Rosaceae (Blackthorn/ Sloe)

*Rhamnus Frangula* f. Rhamnaceae (Alder Buckthorn)

*Rubus fruticosus* f. Rosaceae (Blackberry)

*Salix fragilis* f. Salicaceae (Crack Willow)



**Sambucus ebulus f. Caprifoliaceae (Dwarf Elder)**

**Sambucus nigra f. Caprifoliaceae (Elder)**

**Solanum guineense f. Solanaceae (Garden Huckleberry)**

**Sorindeia werneckei f. Anacardiaceae**

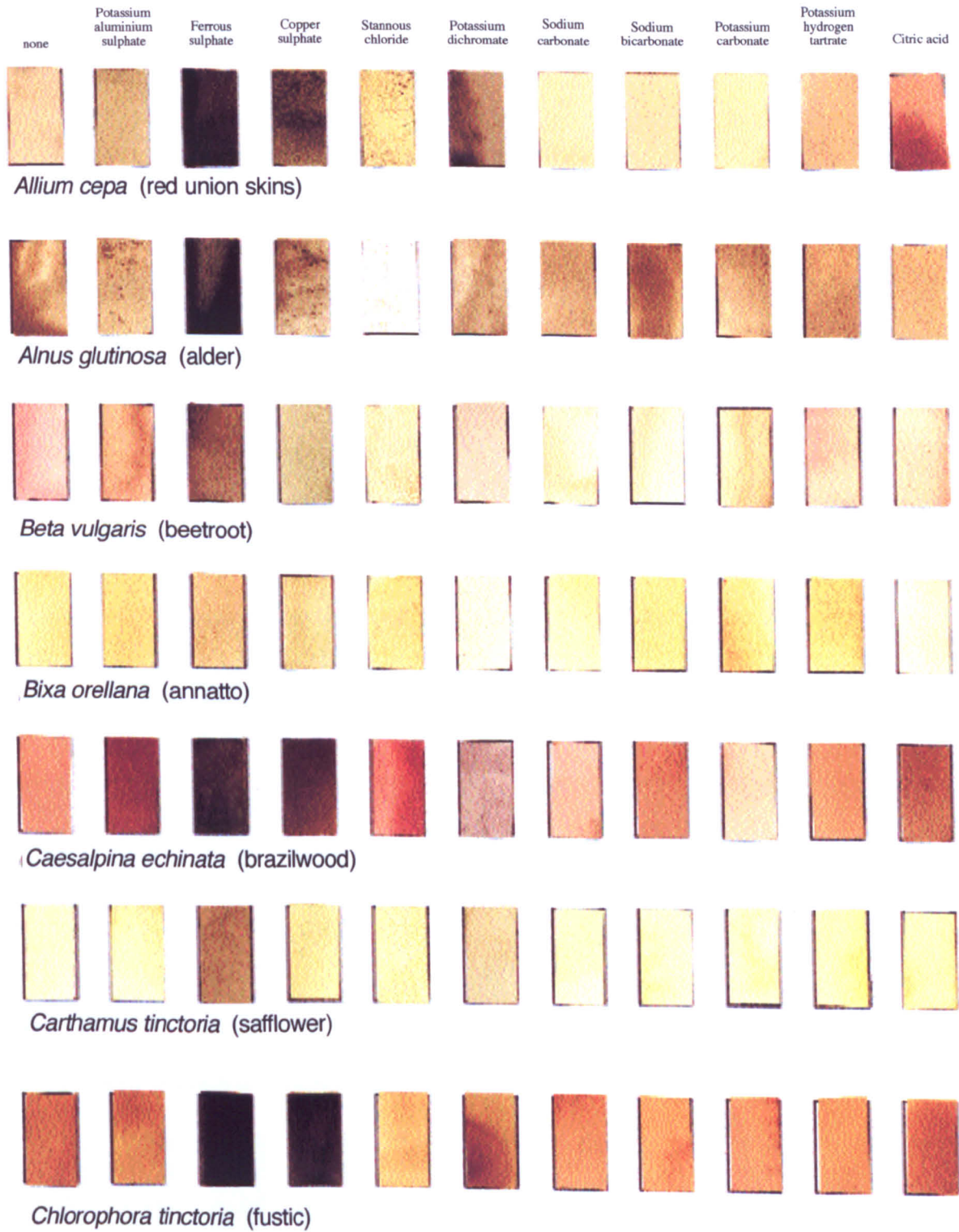
**Uapaca sansibarica f. Euphorbiaceae**

**Wrightia tinc. f. Apocynaceae**

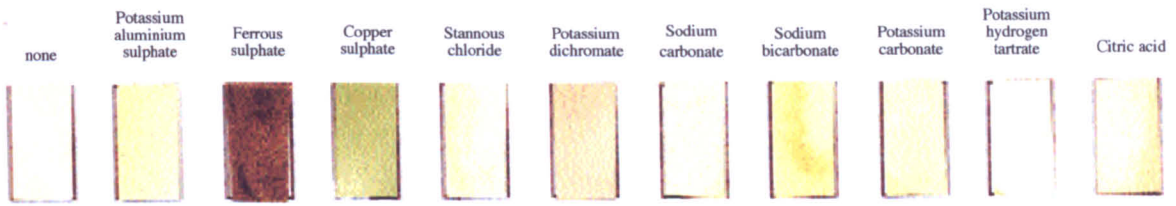


## Appendix 2 Initial Extraction and Modifier Tests (plates)

Of the initial series of 670 tests on 78 species and varieties, 23 of the most promising sources of CMYK colour along with the effects of a range of chemical modifiers are illustrated below, followed by the complete annotated list (appendix 2b).



## Appendix 2 Initial Extraction and Modifier Tests



*Citrus sp.* (orange leaves)



*Coreopsis tinctoria* (coreopsis)



*Haematoxylon campechianum* (logwood)



*Hibiscus sabdariffa* (Jamaican sorrel)



*Indigofera tinctoria* (indigo)



*Mahonia aquifolium* (Oregon grape)



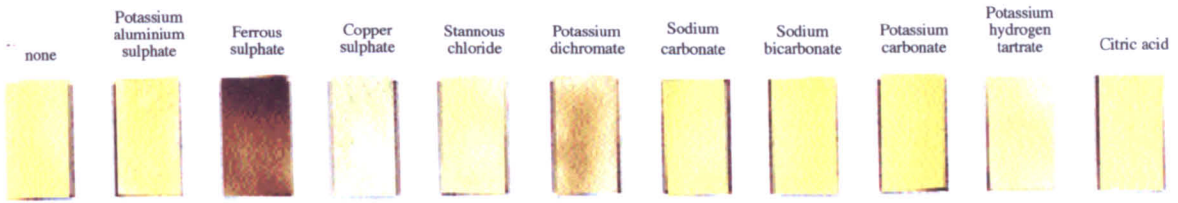
*Phytolacca americana* (pokeweed)



*Prunus laurocerasus* (cherry laurel)



## Appendix 2 Initial Extraction and Modifier Tests



*Reseda luteola* (weld)



*Rhamnus saxatilis* (Persian berry)



*Rubia tinctoria* (madder)



*Rubus fruticosus* (blackberry)



*Sambucus nigra* (elderberry)



*Solanum guineense* (garden huckleberry)



*Vaccinium corymbosum* (blueberry)



Oak galls

## Appendix 2b Initial Extraction and Modifier Tests

The initial series of tests designed to determine the effect of a range of chemical modifiers on plant pigments. Samples were obtained from a variety of sources including the Cat Hill Ink Garden, Chelsea Physic Garden, the Royal Botanical Gardens at Kew, Ashill Colour Studio and a number of field trips in S.E. England.

### Modifiers

As = Potassium aluminium sulphate

Ca = Citric acid

Cr = Potassium dichromate

Bs = Sodium bicarbonate

Cu = Copper sulphate

Pc = Potassium carbonate

Ct = Acid potassium tartrate

Fe = Ferrous sulphate

Sc = Sodium carbonate

Aa = Acetic acid

Sn = Stannous chloride

Oa = Oxalic acid

al = Alcohol (methyl)

Asc = Ascorbic acid

Amo = Ammonia

all temperatures measured in ° centigrade.

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
<i>Aesculus hippocastrum</i>	Hippocastanaceae	nut shells	Ca	sod	100	pale brick	<7	UK
			Cr	sod	100	trans brown	<7	UK
			Bs	sod	100	trans brown	<7	UK
			Cu	sod	100	trans brown	<7	UK
			Pc	sod	100	trans brown	<7	UK
			Ct	sod	100	trans brown	<7	UK
			none	sod	100	trans brown	<7	UK
<i>allium cepa</i>	alliaceae	outer skin	none		100	orange	<7	UK
			Ca	sod	100	pale orange/brown	<7	UK
			As	sod	100	translucent "	<7	UK
			Ct	sod	100	pale orange/brown	<7	UK
			Aa	sod	100	" "	<7	UK
			Cr	sod	100	rich orange/brown	<7	UK
			Cu	sod	100	translucent "	<7	UK
<i>allium cepa (red)</i>	alliaceae	outer skin	Fe	sod	100	mid brown	<7	UK
			none		100	purple	7	UK
			Ct		100	purple	>7	UK
			As		100	viridian	>7	UK
			Aa		100	grey/green	>7	UK
			Ca		100	magenta	>7	UK
			Bs		100	green	<7	UK
<i>allium cepa (red)</i>	alliaceae	outer skin	Fe		100	deep grey/green	>7	UK
			Fe+al		100	deep green	>7	UK
			Fe+Ca		100	maroon	>7	UK
			none	sod	100	green	<7	UK



## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
			Sn	sod	100	v. dark green	<7	UK
			Fe	sod	100	dull green	<7	UK
			As	sod	100	pink	<7	UK
			Ca	sod	100	pale green/brown	<7	UK
			Cr	sod	100	pale brown	<7	UK
			Bs	sod	100	green/brown	<7	UK
			Cu	sod	100	pale ginger/brown	<7	UK
			Pc	sod	100	pale brown	<7	UK
			Ct	sod	100	pale green/brown	<7	UK
			Fe+Ca	sod	100	pale brown	<7	UK
<b>alnus glutinosa</b>	<b>Betulaceae</b>	<b>bark</b>	Sn	sod	100	trans rust	<7	UK
			Fe	sod	100	trans black	<7	UK
			As	sod	100	pale sand	<7	UK
			Ca	sod	100	pale sand	<7	UK
			Cr	sod	100	trans fawn	<7	UK
			Bs	sod	100	trans light brown	<7	UK
			Cu	sod	100	trans fawn	<7	UK
			Pc	sod	100	sand	<7	UK
			Ct	sod	100	light sand	<7	UK
			none	sod	100	trans light brown	<7	UK
<b>Ananas comosus</b>	<b>Bromeliaceae</b>	<b>leaves</b>	none	sod	100	pale yellow	<7	UK
			Cu	sod	100	pale viridian	<7	UK
<b>aster novi belgii</b>	<b>Compositae</b>	<b>whole plant</b>	none		100	pale green	7	UK
<b>Azalea</b>	<b>Ericaceae</b>	<b>leaves</b>	none		100	fawn	7	UK
		<b>leaves</b>	Ct		100	beige	>7	UK
<b>Beta vulgaris</b>	<b>Chenopodiaceae</b>	<b>root</b>	Sn+Asc		100	pale brick	>7	UK
			Fe+Asc		100	pale drab	>7	UK
			as+Asc		100	pale maroon	>7	UK
			Ca+Asc		100	pink	>7	UK
			Cr+Asc		100	v.pale drab	>7	UK
			Bs+Asc		100	light sand	7	UK
			Cu+Asc		100	pale olive	>7	UK
			Pc+Asc		100	light sand	7	UK
			Ct+Asc		100	pale magenta	>7	UK
			Asc		100	" "	>7	UK
			Asc	sod	100	light sand	<7	UK
			none		100	magenta	7	UK
			Cr	sod	100	yellow/green	<7	UK
			Ca		100	magenta	>7	UK
			As		100	magenta	>7	UK
			Aa		100	magenta	>7	UK
<b>Beta vulgaris</b>	<b>Chenopodiaceae</b>	<b>root</b>	Ct		100	magenta	>7	UK
			Fe		100	magenta	>7	UK
			Oa		100	pink	>7	UK
			Cu		100	orange	>7	UK
<b>Betula pendula</b>	<b>Betulaceae</b>	<b>bark</b>	Sn	sod	100	trans mid brown	<7	UK

## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
			Fe	sod	100	dark brown	<7	UK
			As	sod	100	trans mid brown	<7	UK
			Ca	sod	100	trans mid brown	<7	UK
			Cr	sod	100	trans mid brown	<7	UK
			Bs	sod	100	trans mid brown	<7	UK
			Cu	sod	100	trans choc	<7	UK
			Pc	sod	100	trans palebrown	<7	UK
			Ct	sod	100	trans mid brown	<7	UK
			none	sod	100	trans mid brown	7	UK
			none	sod	100	trans mid brown	7	UK
<b>Bixa orellano</b>	<b>Bixaceae</b>	<b>seeds</b>	Sn	sod	100	orange/yellow	<7	S.America
			Fe	sod	100	orange/yellow	<7	S.America
			As	sod	100	orange/yellow	<7	S.America
			Ca	sod	100	orange/yellow	<7	S.America
			Cr	sod	100	orange/yellow	<7	S.America
			Bs	sod	100	orange/yellow	<7	S.America
			Cu	sod	100	orange/yellow	<7	S.America
			Pc	sod	100	orange/yellow	<7	S.America
			Ct	sod	100	orange/yellow	<7	S.America
			none	sod	100	orange/yellow	<7	S.America
			none	sod	100	orange/yellow	<7	S.America
<b>Brassica oleraceae</b>	<b>Crucifereae</b>	<b>leaves</b>	none	sod	100	pale green	<7	UK
<b>Brassica oleraceae (red)</b>	<b>Crucifereae</b>	<b>leaves</b>	Ca	sod	100	purple/violet	<7	UK
			As	sod	100	mid/deep blue	<7	UK
			Ct	sod	100	grey/violet	<7	UK
			Cr	sod	100	mid green	<7	UK
			Fe	sod	100	pale viridian	<7	UK
			Oa	sod	100	mid/dark blue	<7	UK
			CU	sod	100	blue/green	<7	UK
			Ca+As	sod	100	magenta	<7	UK
			Ca+Fe	sod	100	magenta (darker)	<7	UK
			Ca+Aa	sod	100	bright magenta	<7	UK
			Ca+Ct	sod	100	" "	<7	UK
			Ca+Cr	sod	100	brown	<7	UK
			Cax2	sod	100	magenta/verm	<7	UK
		<b>leaves+saffr</b>	none		100	orange	7	UK
<b>Buddleja davidii</b>	<b>Buddlejaceae</b>	<b>d/heads (froz)</b>	Sn	sod	100	trans yell ochre	<7	UK
			Fe	sod	100	" brown	<7	UK
			As	sod	100	" yell ochre	<7	UK
			Ca	sod	100	" sand	<7	UK
			Cr	sod	100	" Khaki	<7	UK
			Bs	sod	100	" yellow	<7	UK
			Cu	sod	100	" green	<7	UK
			Pc	sod	100	" yellow	<7	UK
			Ct	sod	100	v. pale flesh	<7	UK
			none	sod	100	" yell/green	<7	UK

## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
			none	sod	100	yellow	<7	UK
		flowers	none		100	pale yellow	7	UK
		flowers	Aa		100	yellow/brown	>7	UK
		flowers	Ct		100	green/brown	>7	UK
		flowers	Ca	sod	100	yellow ochre	<7	UK
		flowers	BS	sod	100	green/ochre	<7	UK
		flowers	Cu	sod	100	green/ochre	<7	UK
		flowers	Cr	sod	100	green/ochre	<7	UK
		flowers	As	sod	100	green/ochre	<7	UK
		flowers	Fe	sod	100	dark green	<7	UK
<b>Caesalpinia echinata</b>	Leguminosae	bark	As	sod	100	deep red	<7	Brazil
			Ca	sod	100	orange	<7	Brazil
			Cr	sod	100	purp/brown	<7	Brazil
			Bs	sod	100	pale brown	<7	Brazil
			Cu	sod	100	choc brown	<7	Brazil
			Pc	sod	100	pale brown	<7	Brazil
			Ct	sod	100	brick orange/red	<7	Brazil
			none	sod	100	brick red	<7	Brazil
			none	sod	100	mid brown	<7	Brazil
			Sn	sod	100	scarlet	<7	Brazil
			Fe	sod	100	black	<7	Brazil
<b>Calystegia sepium</b>	Convolvulaceae	leaves & buds	none		100	pale ochre	7	UK
<b>Carthamus tinctoria</b>	Compositae	florets	Sn	sod	100	pale yellow	<7	India
			Fe	sod	100	pale yellow	<7	India
			As	sod	100	pale yellow	<7	India
			Ca	sod	100	pale yellow	<7	India
			Cr	sod	100	dirty yellow	<7	India
			Bs	sod	100	mid gold	<7	India
			Cu	sod	100	ochre	<7	India
			Pc	sod	100	pale yellow	<7	India
			Ct	sod	100	v. pale yellow	<7	India
			none	sod	100	gold	<7	India
<b>Chamaecyparis lawsoniana</b>	Cupressae	cones	none	sod	100	red	<7	UK
			Ct	sod	100	pink	<7	UK
			Fe	sod	100	purple/black	<7	UK
<b>Chlorophora tinctoria</b>	Moraceae	wood	Cr	sod	100	rich tile red	<7	UK
			Sn		100	trans lem yellow	>7	Brazil
			Fe		100	" brown	>7	Brazil
			As		100	" chrome yell	>7	Brazil
<b>Chlorophora tinctoria</b>	Moraceae	wood	Ca		100	" pale yellow	>7	Brazil
			Cr		100	brown	>7	Brazil
			Bs		100	pale ginger brown	<7	Brazil
			Cu		100	ochre	>7	Brazil
			Pc		100	pale ginger brown	<7	Brazil
			Ct		100	chrome yellow	>7	Brazil
			none		100	deep chrome yell	7	Brazil

## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
			none	sod	100	pale ginger brown	<7	Brazil
<i>Cichorium intybus</i>	Compositae	leaves	none	sod	100	green	<7	UK
<i>Circuma longa</i>	Zingiberaceae	root	none	sod	100	orange/red	<7	India
			Sn	sod	100	trans orange	<7	India
			Fe	sod	100	mid brown	<7	India
			As	sod	100	yellow	<7	India
			Ca	sod	100	light chrome	<7	India
			Cr	sod	100	khaki	<7	India
			Bs	sod	100	orange	<7	India
			Cu	sod	100	trans yellow/grn	<7	India
			Pc	sod	100	deep orange	<7	India
			Ct	sod	100	lemon yellow	<7	India
			none	sod	100	yellow ochre	<7	India
			none		100	yellow	7	India
<i>Citrus acida</i>	Ruticeae	rind	none	sod	100	deep yellow	<7	UK
<i>Citrus limon</i>	Ruticeae	rind	Ct	sod	100	green/ochre	<7	UK
			Cu	sod	100	lemon yellow	<7	UK
			Cr	sod	100	dark yellow/grn	<7	UK
			Ca	sod	100	pale yellow	<7	UK
			Fe	sod	100	brown	<7	UK
			none	sod	100	orange/yellow	<7	UK
<i>Citrus sinensis</i>	Ruticeae	leaves	none		100	lemon yellow	>7	UK
			As		100	pale yellow	>7	UK
			Cr		100	yellow	7	UK
			Fe		100	brown	>7	UK
<i>Clematis montana</i>	Ranunculaceae	flowers	none		100	pale grey	>7	UK
			Fe		100	blue/grey	>7	UK
			Ct		100	pale grey	>7	UK
			As		100	pale grey/green	>7	UK
			Ca		100	pink	>7	UK
			Cr		100	olive	>7	UK
<i>Clerodendrum trichotomum</i>	Verbenaceae	berry (froz)	Sn		100	v.pale green	>7	UK
			Fe		100	pale brown/green	>7	UK
			As		100	v. pale green	>7	UK
			Ca		100	v. pale green	>7	UK
			Cr		100	v. pale brown/grn	>7	UK
			Bs		100	pale green	<7	UK
			Cu		100	pale blue/green	>7	UK
			Pc		100	pale green	<7	UK
			Ct		100	pale green	>7	UK
			none		100	pale green	7	UK
			none		100	pale green	7	UK
<i>Coreopsis tinctoria</i>	Compositae	flowers	Sn		100	pale orange	>7	UK
			Fe		100	fawn	>7	UK
			As		100	orange	>7	UK
			Ca		100	lemon	>7	UK

## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
			Cr		100	fawn	>7	UK
			Bs		100	orange/vermilion	<7	UK
			Cu		100	sand	>7	UK
			Pc		100	orange	<7	UK
			Ct		100	light chrome yell	>7	UK
			none		100	pale orange	7	UK
			none	sod	100	orange	<7	UK
<b>Cotoneaster horizontalis</b>	<b>Rosaceae</b>	<b>leaves</b>	none		100	deep orange/brwn	7	UK
			none	sod	100	red brown	<7	UK
			Cr	sod	100	rich yellow/brown	<7	UK
			Fe	sod	100	dark choc	<7	UK
<b>Crataegus oxycanthus</b>	<b>Rosaceae</b>	<b>bark</b>	none		100	pink/brown	7	UK
			Fe		100	purple/black	>7	UK
			Cr		100	ginger/brown	>7	UK
			Fe	sod	100	purple	<7	UK
			Cr	sod	100	brown	<7	UK
			none	sod	100	red/brown	<7	UK
<b>Crocus sativus</b>	<b>Iridaceae</b>	<b>anther</b>	none		100	bright yellow	7	Spain
<b>Cucumis melo</b>	<b>Cucurbitaceae</b>	<b>seeds</b>	Bs	sod	100	gold	<7	UK
			Pc	sod	100	gold	<7	UK
<b>Evernia prunasti</b>	<b>Lecanorales</b>	<b>whole</b>	Sn	amo	100	pale blue/grey	<7	UK
			Fe	amo	100	pale blue/grey	<7	UK
			As	amo	100	pale blue/grey	<7	UK
			Ca	amo	100	pale blue/grey	<7	UK
			Cr	amo	100	pale blue/grey	<7	UK
			Bs	amo	100	pale blue/grey	<7	UK
			Cu	amo	100	pale blue/grey	<7	UK
			Pc	amo	100	pale blue/grey	<7	UK
<b>Evernia prunasti</b>	<b>Lecanorales</b>	<b>whole</b>	Ct	amo	100	pale blue/grey	<7	UK
			none	amo	100	pale blue/grey	<7	UK
			none	amo	100	pale blue/grey	<7	UK
<b>Fagus Cuprea</b>	<b>Fagaceae</b>	<b>bark</b>	none		100	brown	7	UK
			Ct		100	brown	>7	UK
			Aa		100	deep green	>7	UK
			Ca	sod	100	orange/brown	<7	UK
<b>Fagus Cuprea</b>	<b>Fagaceae</b>	<b>bark</b>	Fe	sod	100	deep choc brown	<7	UK
			Ct	sod	100	mid red/brown	<7	UK
			Aa	sod	100	mid red/brown	<7	UK
			As	sod	100	mid red/brown	<7	UK
<b>Fagus sylvatica</b>	<b>Fagaceae</b>	<b>bark</b>	none		100	pink/red	7	UK
			Cr		100	ochre	>7	UK
			Fe		100	pale brown	>7	UK
<b>Forsythia ovata</b>	<b>Oleaceae</b>	<b>leaves</b>	Ca		100	dull yellow green	>7	UK
			Ct		100	dull yellow green	>7	UK
			Aa		100	dull yellow green	>7	UK
			Cr		100	dull yellow green	>7	UK

## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>			
<i>Fraxinus excelsior</i>	Oleaceae	leaves	As		100	dull yellow green	>7	UK			
			Fe		100	grey/brown	>7	UK			
			none		100	yellow ochre	7	UK			
			Fe	sod	100	grey	<7	UK			
<i>Fuschia magellanica</i>	Onagraceae	flowers	Oa	sod	100	brown	<7	UK			
			Aa		100	purple/blue	>7	UK			
			none	sod	100	green	<7	UK			
<i>Genista hispanica</i>	Leguminosae	flowers	Sn	sod	100	pale yellow	<7	UK			
			Fe	sod	100	pale mid brown	<7	UK			
			As	sod	100	pale yellow	<7	UK			
			Ca	sod	100	v.pale sand	<7	UK			
			Cr	sod	100	v.pale green	<7	UK			
			Bs	sod	100	pale yellow	<7	UK			
			Cu	sod	100	pale green	<7	UK			
			Pc	sod	100		<7	UK			
			Ct	sod	100	pale yellow	<7	UK			
			none	sod	100	pale sand	<7	UK			
			none	sod	100	pale yellow	<7	UK			
			<i>Glycyrrhiza glabra</i>	Leguminosae	root	Sn	sod	100	trans sand	<7	Spain
						Fe	sod	100	mid brown	<7	Spain
						As	sod	100	trans sand	<7	Spain
Ca	sod	100				trans pale brown	<7	Spain			
Cr	sod	100				fawn	<7	Spain			
Bs	sod	100				gold	<7	Spain			
Cu	sod	100				trans pale olive	<7	Spain			
<i>Glycyrrhiza glabra</i>	Leguminosae	root	Pc	sod	100	gold	<7	Spain			
			Ct	sod	100	pale gold	<7	Spain			
			none	sod	100	pale gold	<7	Spain			
			none	sod	100	gold	<7	Spain			
<i>Haematoxylon campechianum</i>	Leguminosae	wood	Sn		100	purple/maroon	>7	Haiti			
			Fe		100	black	>7	Haiti			
			As		100	purple	>7	Haiti			
			Ca		100	orange/brown	>7	Haiti			
<i>Haematoxylon campechianum</i>	Leguminosae	wood	Cr		100	pale grey	>7	Haiti			
			Bs		100	light brown	<7	Haiti			
			Cu		100	blue/black	>7	Haiti			
			Pc		100	deep fawn	<7	Haiti			
			Ct		100	mid red/brown	>7	Haiti			
			none		100	choc brown	7	Haiti			
			none	sod	100	light brown	<7	Haiti			
			none		100	pale blue/green	7	UK			
<i>Helianthus annuus</i>	Compositae	seeds	none	sod	100	deep green	<7	UK			
			Fe	sod	100	choc brown	<7	UK			
			Cr	sod	100	green/brown	<7	UK			
			As	sod	100	pale green	<7	UK			
			Ct	sod	100	mid green	<7	UK			

## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
			Aa	sod	100	mid-dark green	<7	UK
			Bs	sod	100	deep green	<7	UK
			Cu	sod	100	mid blue/green	<7	UK
<b>Hibiscus sabdariffa</b>	<b>Malvaceae</b>	<b>fruit</b>	Sn		100	trans. violet	>7	Jamaica
			Fe		100	dark sand	>7	Jamaica
			As		100	trans purp/pink	>7	Jamaica
			Ca		100	trans pink	>7	Jamaica
			Cr		100	grey	>7	Jamaica
			Bs		100	trans purple	<7	Jamaica
			Cu		100	trans pale maroon	>7	Jamaica
			Pc		100	trans pale turq	<7	Jamaica
			Ct		100	trans deep pink	>7	Jamaica
			none		100	trans pale pink	7	Jamaica
<b>Hydrangea macrophylla</b>	<b>Hydraneaceae</b>	<b>flowers</b>	none		100	brown/ochre	7	UK
				sod	100	pale ochre	<7	UK
			Cr	sod	100	pale ochre	<7	UK
			As	sod	100	pale ochre	<7	UK
			Fe	sod	100	mid brown	<7	UK
<b>Hypericum elatum</b>	<b>Guttifereae</b>	<b>ripe berry</b>	none	sod	100	mid brown	<7	UK
			Cr	sod	100	choc brown	<7	UK
			Fe	sod	100	brown	<7	UK
			Ct	sod	100	light brown	<7	UK
<b>Hypericum elatum</b>	<b>Guttifereae</b>	<b>ripe berry</b>	Aa	sod	100	light brown	<7	UK
			Ca	sod	100	light brown	<7	UK
			As	sod	100	trans brown	<7	UK
			Cu	sod	100	trans brown	<7	UK
<b>Ilex crenata</b>	<b>Aquifoliaceae</b>	<b>leaves</b>	none		100	golden brown	7	UK
<b>Indigofera tinctoria</b>	<b>Leguminosae</b>	<b>leaves</b>	Sn	amo	100	Blue/grey	<7	India
			Fe	amo	100	dark green	<7	India
			As	amo	100	blue/grey	<7	India
			Ca	amo	100	blue/grey	<7	India
<b>Indigofera tinctoria</b>	<b>Leguminosae</b>	<b>leaves</b>	Cr	amo	100	khaki	<7	India
			Bs	amo	100	blue/grey	<7	India
			Cu	amo	100	blue/grey	<7	India
			Pc	amo	100	blue/grey	<7	India
			Ct	amo	100	blue/grey	<7	India
			none	amo	100	blue/grey	<7	India
			none	amo	100	blue/grey	<7	India
<b>Iris foetidissima</b>	<b>Iridaceae</b>	<b>berry (froz)</b>		sod	100	light brown	<7	UK
<b>Juniperus communis</b>	<b>Cupressaceae</b>	<b>dried berry</b>		sod	100	pale sand	<7	UK
<b>Laburnum vulgare</b>	<b>Leguminosae</b>	<b>leaves</b>	none		100	dark yellow	7	UK
			none		100	ochre	7	UK
<b>Lavatera maritima</b>	<b>Malvaceae</b>	<b>leaves</b>	none		100	pale yellow	7	UK
<b>Lawsonia inermis/alba</b>	<b>Lythraceae</b>	<b>leaves</b>	none	sod	100	pale gold	<7	India
			Sn	sod	100	dark brown	<7	India
			Fe	sod	100	deep gold	<7	India

## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
			As	sod	100	pale fawn	<7	India
			Ca	sod	100	mid/light brown	<7	India
			Cr	sod	100	orange	<7	India
			Bs	sod	100	fawn	<7	India
			Cu	sod	100	pale orange	<7	India
			Pc	sod	100	pale apricot	<7	India
			Ct	sod	100	trans apricot	<7	India
			none		100	rust	7	India
			none	sod	100	trans pale brown	<7	India
		leaves 3wk sk	Sn	sod	100	light brown	<7	India
		leaves 3wk sk	Fe	sod	100	dark brown	<7	India
		leaves 3wk sk	As	sod	100	ginger brown	<7	India
		leaves 3wk sk	Ca	sod	100	pale ging. brown	<7	India
		leaves 3wk sk	Cr	sod	100	light brown	<7	India
		leaves 3wk sk	Bs	sod	100	rich ging. brown	<7	India
		leaves 3wk sk	Cu	sod	100	trans fawn	<7	India
		leaves 3wk sk	Pc	sod	100	ginger brown	<7	India
		leaves 3wk sk	Ct	sod	100	trans ging brown	<7	India
		leaves 3wk sk	none	soda	100	fawn	<7	India
<b>Lawsonia inermis/alba</b>	<b>Lythraceae</b>	leaves 3wk sk	soda	soda	100	ginger brown	<7	India
<b>Ligustrum japonicum</b>	<b>Oleaceae</b>	leaves	none		100	dark grey	7	UK
<b>Ligustrum vulgare</b>	<b>Oleaceae</b>	berry (froz)	Sn		100	pale ochre	>7	UK
			Fe		100	pale khaki	>7	UK
			As		100	pale brown	>7	UK
			Ca		100	pale ginger	>7	UK
			Cr		100	pale brown/grn	>7	UK
			Bs		100	sand	<7	UK
			Cu		100	fawn	>7	UK
			Pc		100	v.pale ochre	<7	UK
<b>Ligustrum vulgare</b>	<b>Oleaceae</b>	berry (froz)	Ct		100	pale brown	>7	UK
			none		100	" "	7	UK
<b>Lycopersicon esculentum</b>	<b>Solanaceae</b>	leaves	none	sod	100	pale dirty green	<7	UK
			Aa	sod	100	dark dirty green	<7	UK
<b>Pelargonium (zonal)</b>	<b>Geraniaceae</b>	dead heads	none	sod	100	grey/green	<7	UK
			Fe	sod	100	dark grey/brown	<7	UK
			Ca+Cr	sod	100	sand	<7	UK
			Cs	sod	100	translucent brown	<7	UK
			As	sod	100	translucent grey	<7	UK
			Ca	sod	100	brick red	<7	UK
			Aa	sod	100	pale grey	<7	UK
			Ct	sod	100	pale grey/brown	<7	UK
			Cr	sod	100	yellow/green	<7	UK
<b>Persea americana</b>	<b>Lauraceae</b>	skins	Ca	sod	100	deep red/brown	<7	UK
			Cr	sod	100	choc brown	<7	UK
			Fe	sod	100	purp/brown	<7	UK
<b>Phytolacca americana</b>	<b>Phytolaccaceae</b>	berry	Sn		100	pale magenta	>7	UK



## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
			Fe		100	brown	>7	UK
			As		100	purple	>7	UK
			Ca		100	magenta	>7	UK
			Cr		100	khaki	>7	UK
			Bs		100	v. pale khaki	<7	UK
			Cu		100	ginger/tan	>7	UK
			Pc		100	v. pale olive	<7	UK
			Ct		100	bright magenta	>7	UK
			none		100	" "	7	UK
			none	sod	100	pale maroon	<7	UK
<b>Picris echioides</b>	Compositae	dead heads	none		100	ochre/green	n	UK
<b>Prunus domestica</b>	Rosaceae	fruit skins	none		100	plum red	>7	UK
			Fe		100	purple	>7	UK
			Cr		100	khaki	>7	UK
			As		100	pale plum red	>7	UK
			Ct		100	mauve	>7	UK
<b>Prunus domestica</b>	Rosaceae	fruit skins	Ca		100	pink/red	>7	UK
			Cr+Fe		100	green/blue	>7	UK
			Cs		100	deep blue	>7	UK
<b>Prunus laurocerasus</b>	Rosaceae	berry (froz)	Sn		100	pale pink/grey	>7	UK
			Fe		100	pale olive	>7	UK
			As		100	pale grey	>7	UK
			Ca		100	pink	>7	UK
			Cr		100	pale green	>7	UK
			Bs		100	v. pale fawn	<7	UK
			Cu		100	mid brown	>7	UK
			Pc		100	pale fawn	<7	UK
<b>Prunus laurocerasus</b>	Rosaceae	berry (froz)	Ct		100	pink/brown	>7	UK
			none		100	pale pink/brown	>7	UK
			none	sod	100	brown	<7	UK
		berry	Sn		100	trans brown	>7	UK
		berry	Fe		100	deep khaki	>7	UK
		berry	As		100	pale choc	>7	UK
		berry	Ca		100	pale ginger brown	>7	UK
		berry	Cr		100	ochre	>7	UK
		berry	Bs		100	light brown	<7	UK
		berry	Cu		100	mid brown	>7	UK
		berry	Pc		100	light brown	<7	UK
		berry	Ct		100	mid brown	>7	UK
		berry	none		100	pink	>7	UK
		berry	none		100	deep magenta	>7	UK
		berry	Ct+Ca		100	mid magenta	>7	UK
		berry	Ca		100	pale purple	>7	UK
		berry	Ct		100	mid purple	>7	UK
<b>Pterocarpus santalinus</b>	Leguminosae	wood	Sn	sod	100	trans pink	<7	india
			Fe	sod	100	trans mid>brown	<7	India

## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
<i>Reseda luteola</i>	Resedaceae	whole	As	sod	100	pink	<7	India
			Ca	sod	100	pale flesh	<7	India
			Cr	sod	100	pale purp/brown	<7	India
			Bs	sod	100	pale purp/brown	<7	India
			Cu	sod	100	trans " "	<7	India
			Pc	sod	100	pale purp/brown	<7	India
			Ct	sod	100	pale maroon	<7	India
			none	sod	100	dull magenta	<7	India
			none	sod	100	pale purp/brown	<7	India
			Sn	sod	100	pale lemon yellow	<7	Germany
<i>Reseda luteola</i>	Resedaceae	whole	Fe	sod	100	trans fawn	<7	Germany
			As	sod	100	trans pale chrome	<7	Germany
			Ca	sod	100	pale yellow	<7	Germany
			Cr	sod	100	trans dull yellow	<7	Germany
			Bs	sod	100	mid yellow	<7	Germany
			Cu	sod	100	pale trans green	<7	Germany
			Pc	sod	100	mid yellow	<7	Germany
			Ct	sod	100	pale trans yellow	<7	Germany
			none	sod	100	mid yellow	<7	Germany
			none	sod	100	dull yellow	<7	Germany
<i>Rhamnus saxatilis</i>	Rhamnaceae	unripe berry	Sn	sod	100	acid yellow	<7	Croatia
			Fe	sod	100	dark olive	<7	Croatia
			As	sod	100	dull yellow	<7	Croatia
			Ca	sod	100	v. pale ochre	<7	Croatia
			Cr	sod	100	trans brown	<7	Croatia
			Bs	sod	100	chrome yellow	<7	Croatia
			Cu	sod	100	greenish yellow	<7	Croatia
			Pc	sod	100	pale yellow	<7	Croatia
			Ct	sod	100	v. pale yellow	<7	Croatia
			none	sod	100	" "	<7	Croatia
<i>Rhus glabra</i>	Anacardaceae	fruit	none	sod	100	pale yellow	<7	Croatia
			Sn			grey/pink	>7	UK
			Fe			grey	>7	UK
			As			dull pink	>7	UK
			Ca			pale yellow	>7	UK
			Cr			brown	>7	UK
			Bs			brown	<7	UK
			Cu			pale brown	>7	UK
			Pc			brown	<7	UK
			Ct			dull pink	>7	UK
<i>Rubia tinctoria</i>	Rubiaceae	root(24hr sk)	none			pale pink	>7	UK
			Sn	amo	65	pale flesh	<7	Iran
			Fe	amo	65	mid flesh	<7	Iran
			As	amo	65	pink flesh	<7	Iran
			Ca	amo	65	yell flesh	<7	Iran
			Cr	amo	65	pale gold	<7	Iran

## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
			Bs	amo	65	deep pink	<7	Iran
			Cu	amo	65	trans beige	<7	Iran
			Pc	amo	65	pale pink	<7	Iran
			Ct	amo	65	pink	<7	Iran
			none	amo	65	pale flesh	<7	Iran
			none	amo	65	pink	<7	Iran
			none	al	65	deep pink	>7	Iran
<b>Rubus fruticososa</b>	Rosaceae	berry	none		100	orange/brown	>7	UK
			none		100	purple/magenta	>7	UK
			Ca		100	magenta	>7	UK
<b>Rubus fruticososa</b>	Rosaceae	berry	Ct		100	magenta/maroon	>7	UK
<b>Salix babylonica</b>	Salicaceae	bark	none		100	pink/brown	7	UK
			Ba		100	pale brown	>7	UK
			As		100	v. pale brown	>7	UK
			Bs		100	ginger brown	<7	UK
			Ct		100	pale brown	>7	UK
			Fe	sod	100	purple/brown	<7	UK
			Aa	sod	100	red/brown	<7	UK
			Cu	sod	100	trans brown	<7	UK
			Cr	sod	100	mid brown	<7	UK
			As	sod	100	trans brown	<7	UK
			Ct	sod	100	pink/brown	<7	UK
<b>Sambucus nigra</b>	Caprifoliaceae	unripe berries	Ct		100	ochre	>7	UK
<b>Sambucus nigra</b>	Caprifoliaceae	unripe berries	none		100	pink	>7	UK
			Aa		100	purple/violet	>7	UK
			Ct		100	dull purple	>7	UK
			Aa+Ca		100	bright purp/violet	>7	UK
			Cu		100	deep blue	>7	UK
			Cux2		100	green/blue	>7	UK
			Cr		100	green/blue	>7	UK
			Crx2		100	black	>7	UK
			As		100	violet indigo	>7	UK
			Ca		100	red/purple	>7	UK
			Ct		100	blue/purple	>7	UK
			Fe		100	deep blue/purple	>7	UK
		unripe berry	none	sod	100	ochre	<7	UK
			Aa		100	ochre	>7	UK
			As		100	ochre	>7	UK
			Fe	sod	100	mid brown	<7	UK
			Cr	sod	100	yellow	<7	UK
			Cu	sod	100	leaf green	<7	UK
		berry 12wk sk	Sn		100	sand	>7	UK
			Fe		100	sand	>7	UK
			As		100	sand	>7	UK
			Ca		100	light brown	>7	UK

## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
			Cr		100	sand	>7	UK
			Bs		100	sand	<7	UK
			Cu		100	pale olive	>7	UK
			Pc		100	sand	<7	UK
			Ct		100	sand	>7	UK
			none		100	sand	>7	UK
			none	sod	100	sand	<7	UK
		berry (froz)	Sn		100	deep purple	>7	UK
<b>Sambucus nigra</b>	Caprifoliaceae	berry (froz)	Fe		100	blue/black	>7	UK
			As		100	purple	>7	UK
			Ca		100	maroon/magenta	>7	UK
			Cr		100	dirty green	>7	UK
			Bs		100	lovat	<7	UK
			Cu		100	purple/brown	>7	UK
			Pc		100	light ginger	<7	UK
			Ct		100	maroon	>7	UK
			none		100	maroon	>7	UK
			none	sod	100	brown	<7	UK
<b>Solanum melongena</b>	Solanaceae	skins	none	sod	100	pale brown	<7	UK
		bark	none		100	pale brown	7	UK
<b>Solanum scabrum/intrusum</b>	Solanaceae	berry	Sn		100	royal blue	>7	UK
			Fe		100	dark green	>7	UK
<b>Solanum scabrum/intrusum</b>	Solanaceae	berry	As		100	ultramarine	>7	UK
			Ca		100	purple	>7	UK
			Cr		100	pale ochre	>7	UK
			Bs		100	pale green	<7	UK
			Cu		100	prussian blue	>7	UK
			Pc		100	pale green	<7	UK
			Ct		100	pale purple	>7	UK
			none		100	pale purple	>7	UK
			none	sod	100	pale blue/green	<7	UK
<b>Symphoricarpos rivularis</b>	Caprifoliaceae	berry	none		100	yellow/green	>7	UK
			none	sod	100	dark yellow	<7	UK
			Cr	sod	100	lemon	<7	UK
<b>Tagetes patula</b>	Compositae	dead heads	none	sod	100	pale ochre	<7	UK
			Aa	sod	100	pale ochre	<7	UK
			Cu	sod	100	translucent ochre	<7	UK
			As	sod	100	sand	<7	UK
			Cr	sod	100	dark sand	<7	UK
			Fe	sod	100	grey	<7	UK
<b>Taxus baccata</b>	Taxaceae	leaves/stalks	none		100	ginger/brown	7	UK
<b>Tulipa darwinii ('black')</b>	Liliaceae	flower	Sn		100	pale fawn	>7	Holland
			Fe		100	" "	>7	Holland
			As		100	" "	>7	Holland
			Ca		100	pink	>7	Holland
			Cr		100	pale khaki	>7	Holland

## Appendix 2b Initial Extraction and Modifier Tests

<i>plant name</i>	<i>family</i>	<i>part</i>	<i>mod</i>	<i>add</i>	<i>temp</i>	<i>colour</i>	<i>pH</i>	<i>Origin</i>
			Bs		100	pale lime green	<7	Holland
			Cu		100	drab	>7	Holland
			Pc		100	pale yellow	<7	Holland
			Ct		100	v. pale maroon	>7	Holland
			none		100	pale olive	7	Holland
			none	sod	100	dirty green	<7	Holland
			Sn		100	transpurp	>7	Holland
<i>Vaccinium corymbosum</i>	Ericaceae	berry	Fe		100	trans purp/black	>7	UK
		berry	As		100	trans deep purp	>7	UK
		berry	Ca		100	trans dull magenta	>7	UK
		berry	Cr		100	dirty green/brown	>7	UK
		berry	Bs		100	blue/purp	<7	UK
		berry	Cu		100	deep purp	>7	UK
		berry	Pc		100	pale sand	<7	UK
		berry	Ct		100	trans purp	>7	UK
		berry	none		100	trans purp	>7	UK
		berry	none		100	trans blue/grey	>7	UK
<i>Viburnum davidii</i>	Caprifoliaceae	leaves	none	sod	100	orange/brown	<7	UK
		leaves	none		100	pink/brown	>7	UK



### **Appendix 3 Chief Sources of CMYK Colour.**

The following plates illustrate the major plant sources used in the production of the inks described in the study. Unless otherwise stated all photographs were taken by the author.

### Appendix 3 Chief Sources of CMYK Colour.



*Indigofera tinctoria*

Young plant grown by the author.



Woad (*Isatis tinctoria*) - first year growth

University of Tel Aviv Botanical Gardens. Israel.

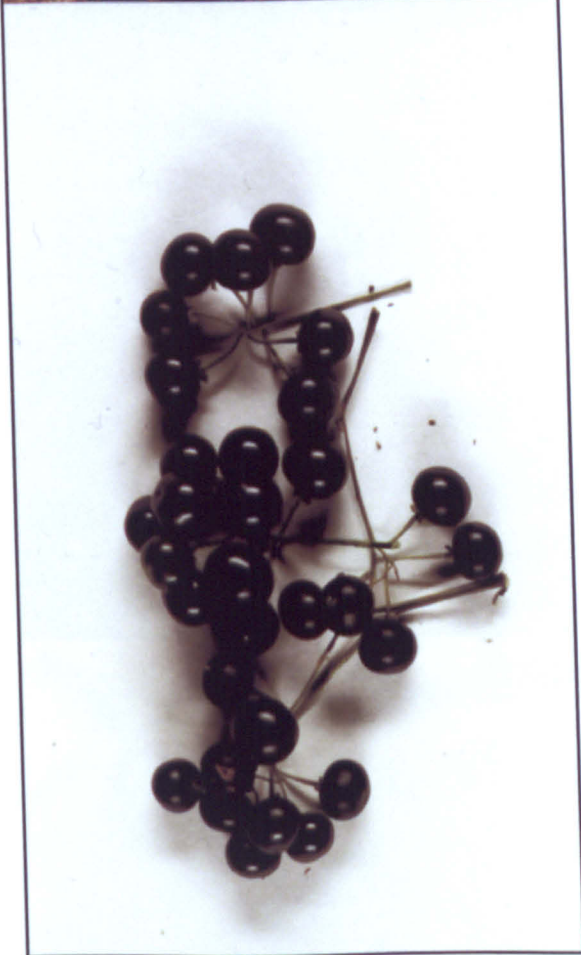


Woad - second year growth

Cat Hill Ink Garden, Middlesex University, UK.



Appendix 3 Chief Sources of CMYK Colour.



Garden Huckleberry (*Solanum guineense*)  
Cat Hill Ink Garden, Middlesex University, UK.

Appendix 3 Chief Sources of CMYK Colour.



Madder (*Rubia tinctoria*)  
Cat Hill Ink Garden, Middlesex University, UK.



Wild madder (*Rubia peregrina*)  
Royal Botanical Gardens, Kew.



Madder root  
Obtained from Dharma Supplies Inc. San  
Raphael, CA. USA.



Appendix 3 Chief Sources of CMYK Colour.



Brazilwood (*Caesalpinia echinata*)

Strybing Arboretum and Botanical Gardens  
San Francisco, USA.



Rasped brazilwood

Obtained from Joseph Flach and Sons Ltd.  
Peterborough, UK.

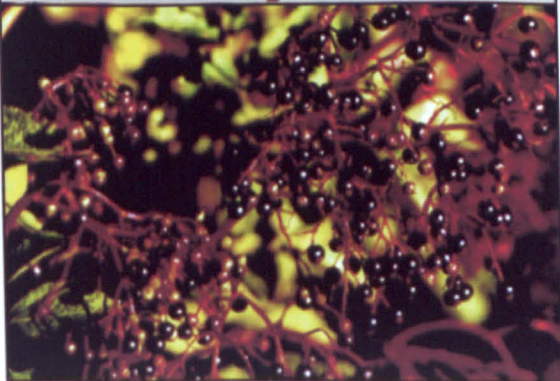
Appendix 3 Chief Sources of CMYK Colour.



Pokeweed  
(*Phytolacca americana*, syn. *P. decandra*)  
Cat Hill Ink Garden, Middlesex University, UK.



Fully ripe pokeberries  
Middlesex University, UK.



Elderberry (*Sambucus nigra*)  
Stoke-by-Nayland, Essex, UK.



Appendix 3 Chief Sources of CMYK Colour.



*Rhamnus saxatilis* ssp. *tinctorius*  
Royal Botanical Gardens, Kew,



Ripe fruit of *Rhamnus saxatilis*  
ssp. *tinctorius*  
Royal Botanical Gardens, Kew,



Persian Berries (dried unripe fruit of  
*Rhamnus saxatilis*)  
Obtained from Ashill Colour Studio, Shefferton, UK.

### Appendix 3 Chief Sources of CMYK Colour.



Weld (*Reseda luteola*)

'Emma's Tradgard', Grafikens Hus. Mariefred Sweden.  
Photograph by Thomas Poranthe



Weld - dried flower heads

Cat Hill Ink Garden, Middlesex University, UK.



Dried and chopped weld

Obtained from Ashill Colour Studio, Shefferton, UK.

**Appendix 3** Chief Sources of CMYK Colour.



Oak Galls  
Stoke-by-Nayland, Essex, UK.



**Appendix 3** Chief Sources of CMYK Colour.



Logwood (*Haematoxylon campechianum*)  
Xoaca, Mexico.



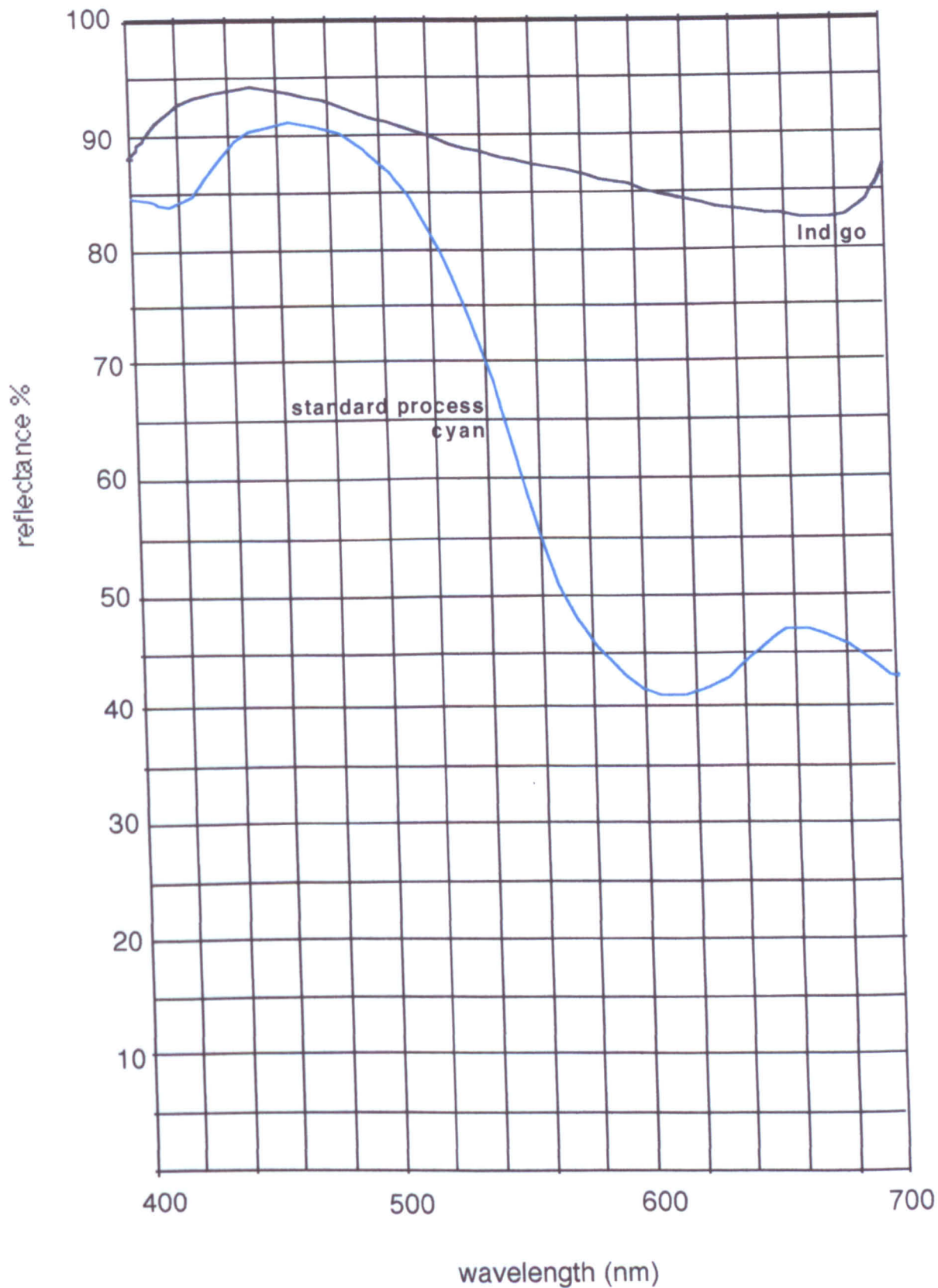
Logwood  
Camp Perrin, Haiti. Photograph Dan Skean



Powdered logwood extract  
Obtained from Joseph Flach and Sons Ltd. Peterborough, UK.

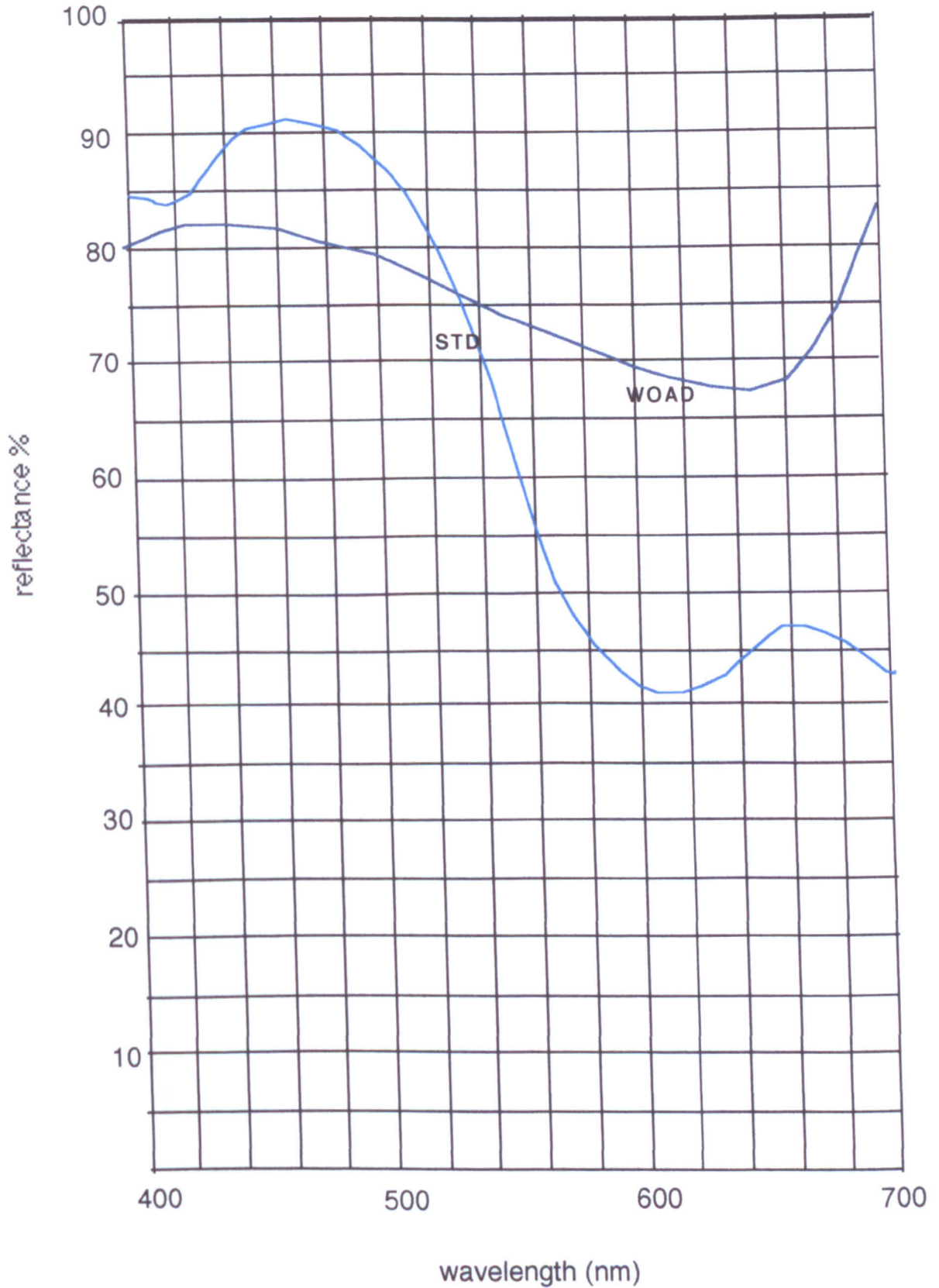


## Appendix 4 Spectral Reflection Comparisons



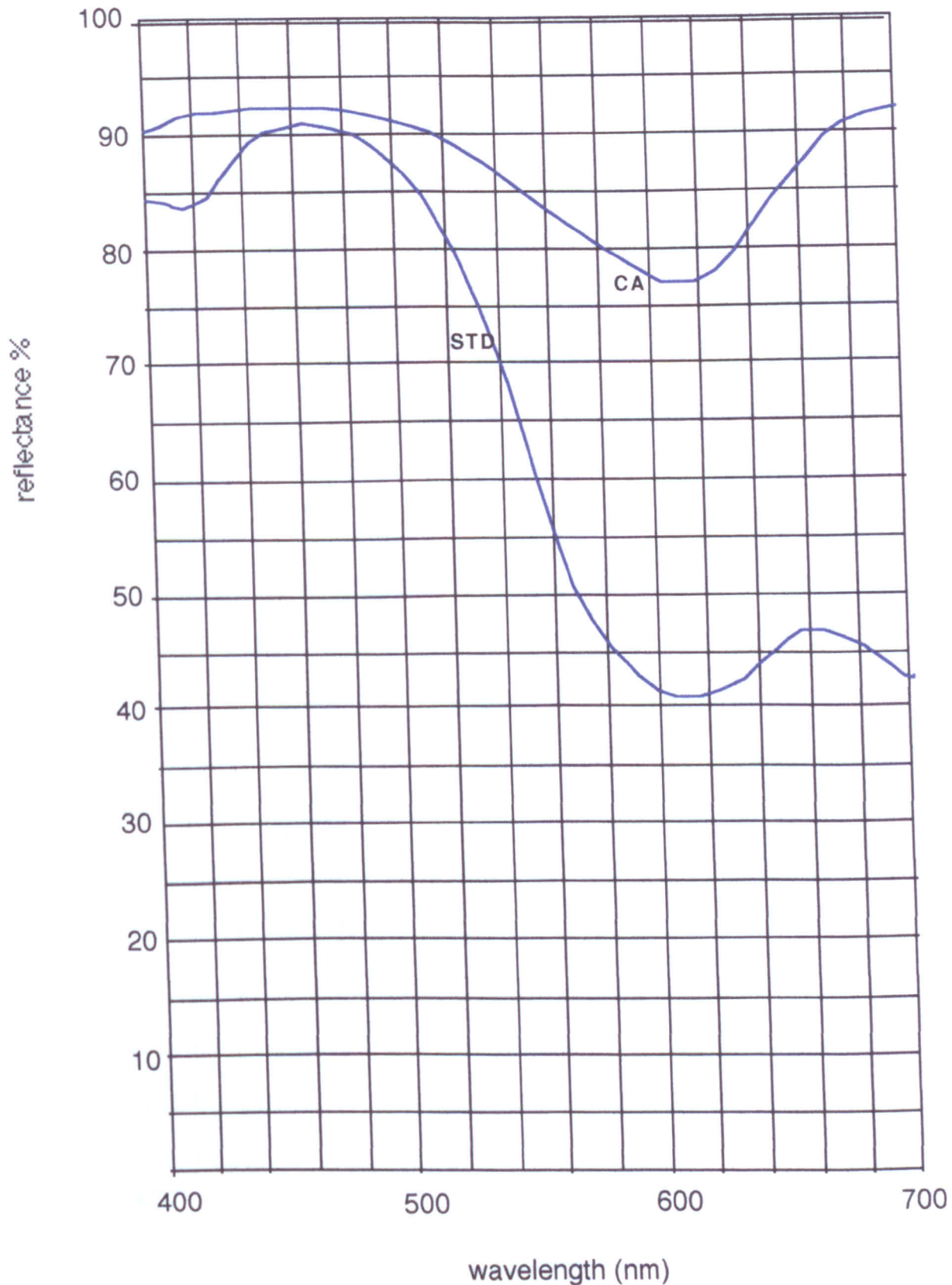
**Natural Indigo.** Natural Indigo powder obtained from a commercial supplier and suspended in sodium alginate.

## Appendix 4 Spectral Reflection



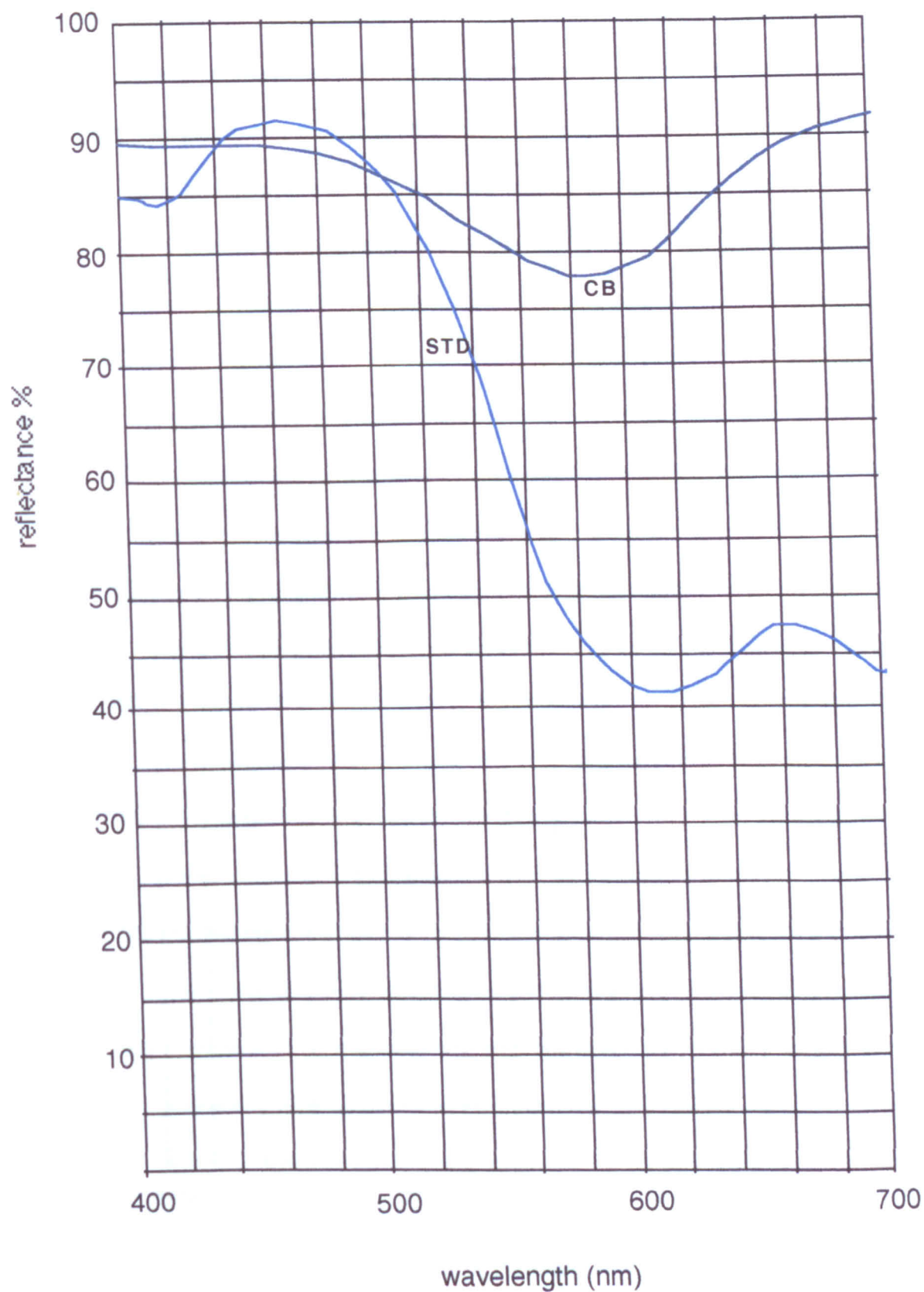
**Woad.** 200gm woad leaves heated in 1000 ml water to 100°C, 10 gm sodium carbonate added to the filtered liquor and aerated to form Indigo precipitate. Suspended in sodium alginate.

## Appendix 4 Spectral Reflection Comparisons



**CA. Sulphonated Indigo Lake.** Prepared with 1 part powdered indigo in 5 parts concentrated sulphuric acid, mixed and neutralised with precipitated chalk. The soluble indigo precipitated onto aluminium hydroxide and suspended in sodium alginate and gum arabic.

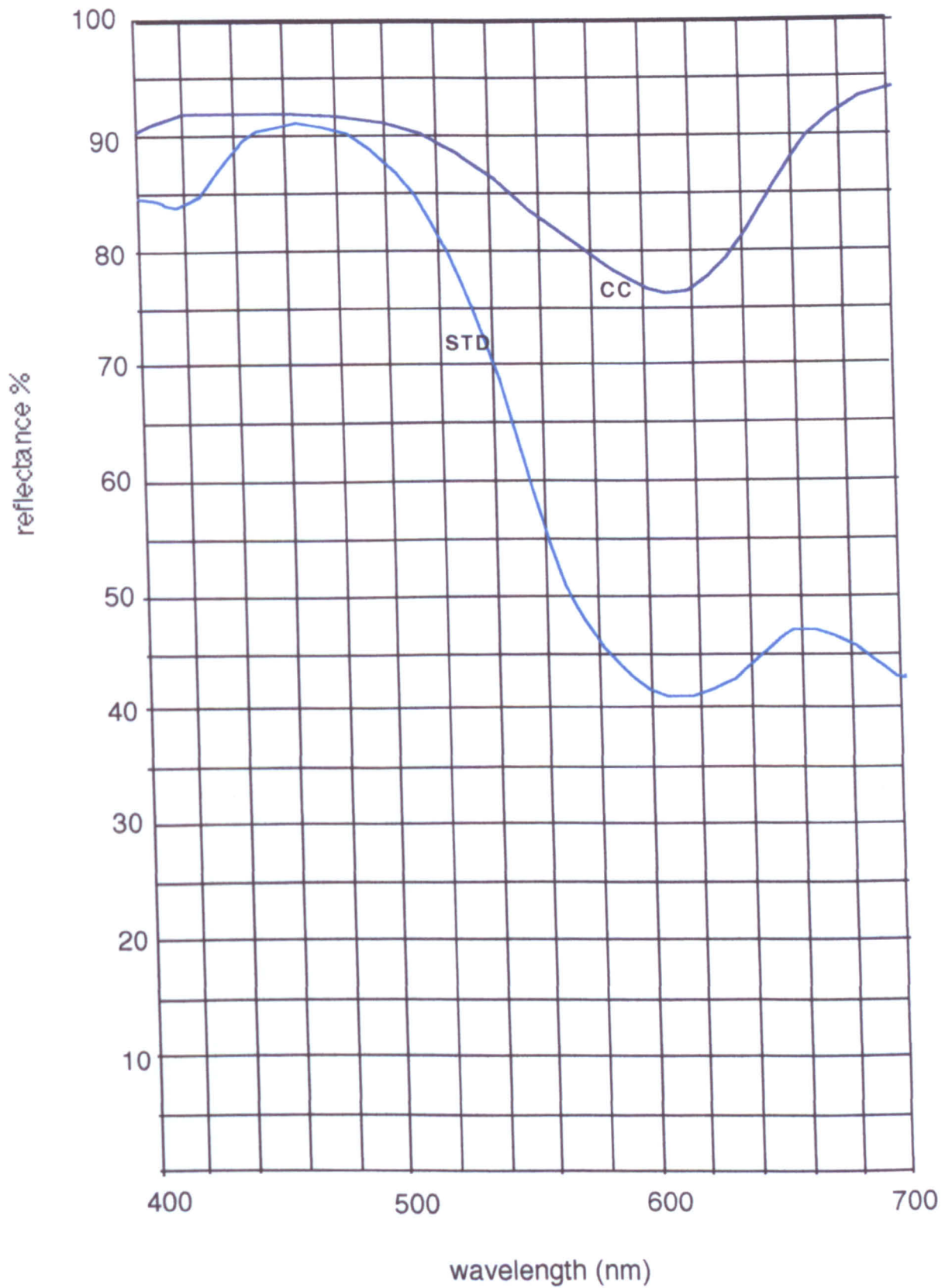
## Appendix 4 Spectral Reflection Comparisons



**CB. Sulphonated Indigo Lake.** Prepared as above and suspended in Rowney System 3 acrylic print medium (6 gm lake/25 ml medium).

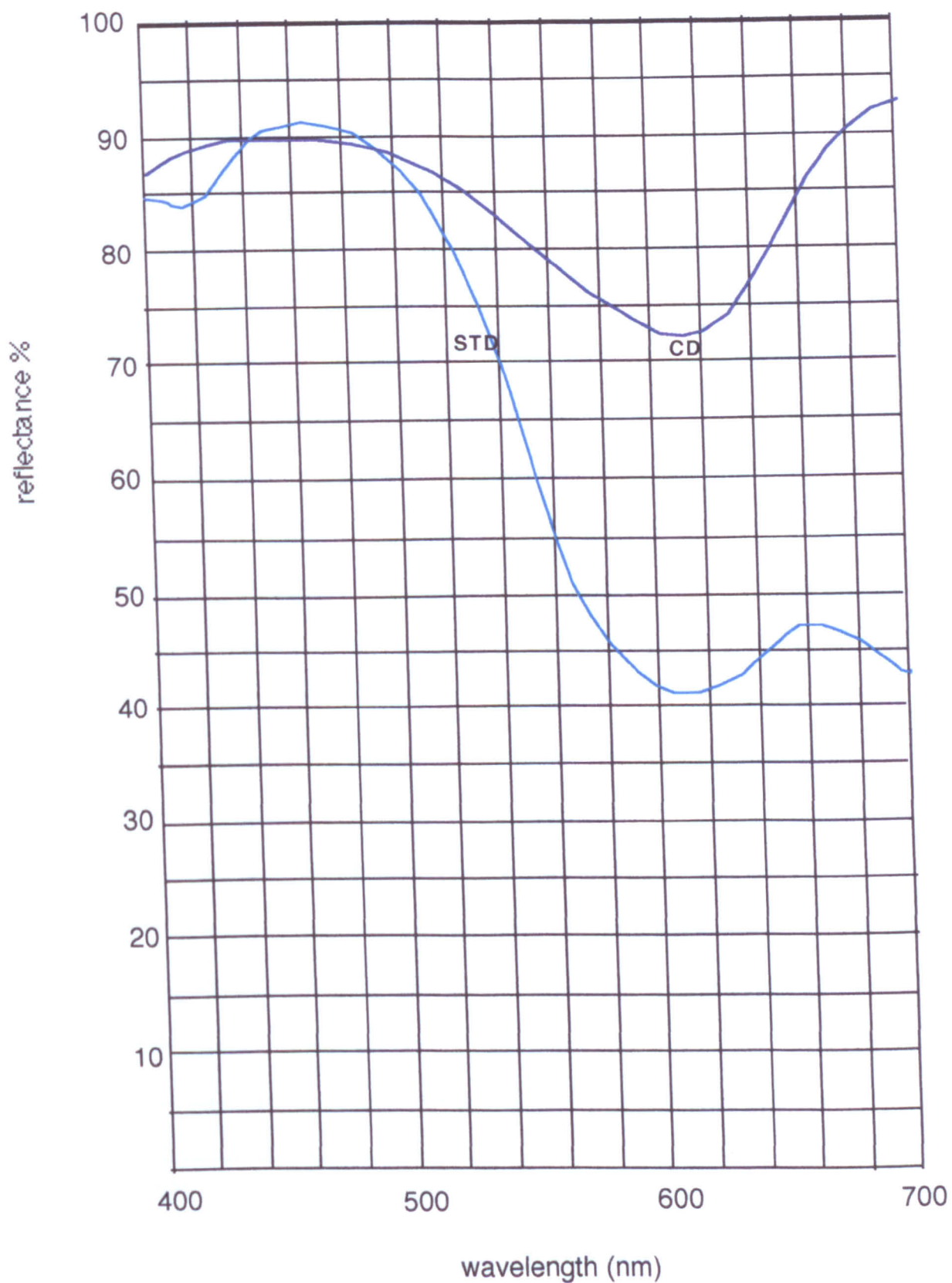


## Appendix 4 Spectral Reflection Comparisons



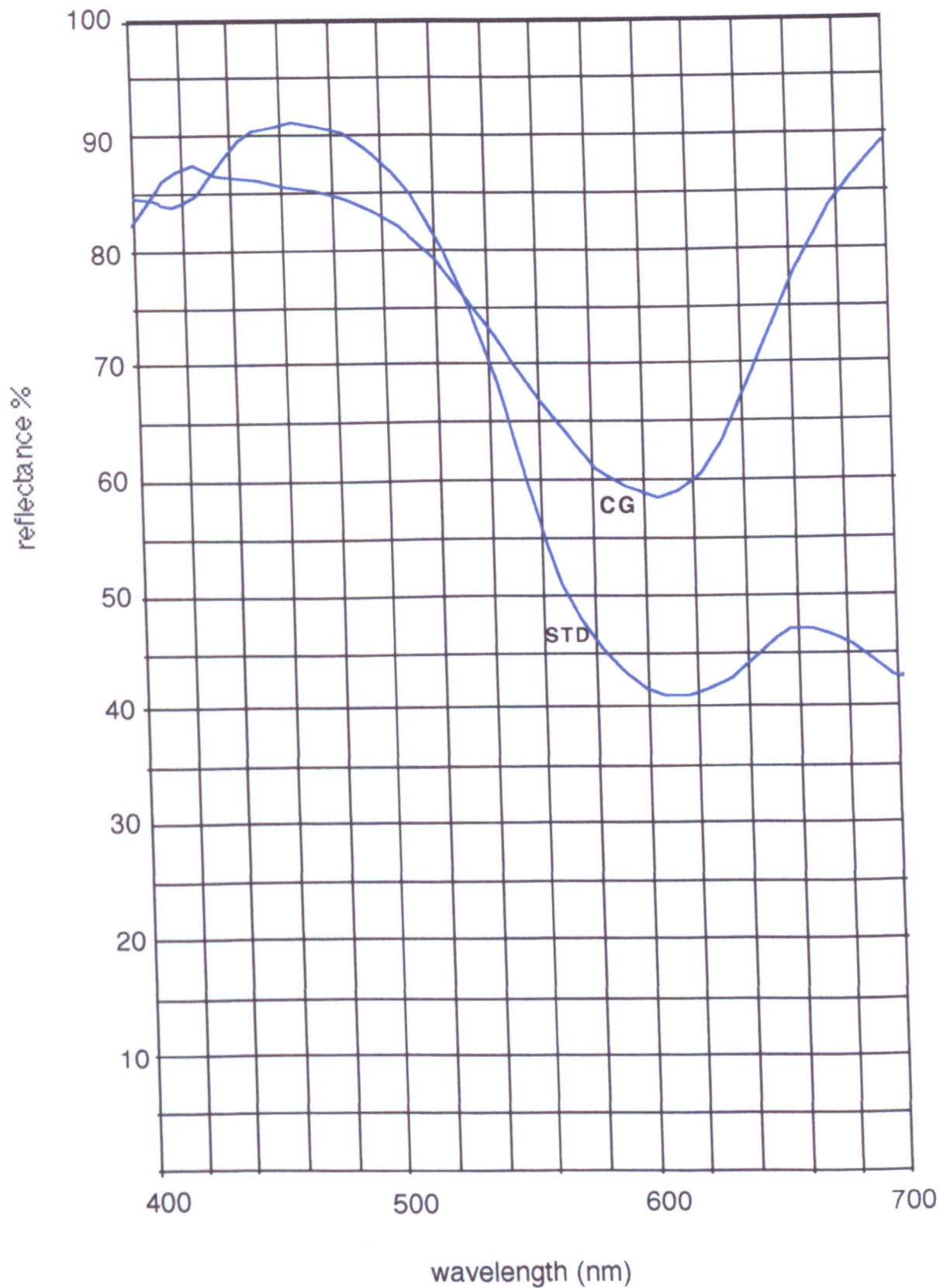
**CC. Sulphonated Indigo Lake.** Prepared as CA and suspended in Rowney System 3 acrylic print medium (12 gm lake/25 ml medium. 2 pulls.

## Appendix 4 Spectral Reflection Comparisons



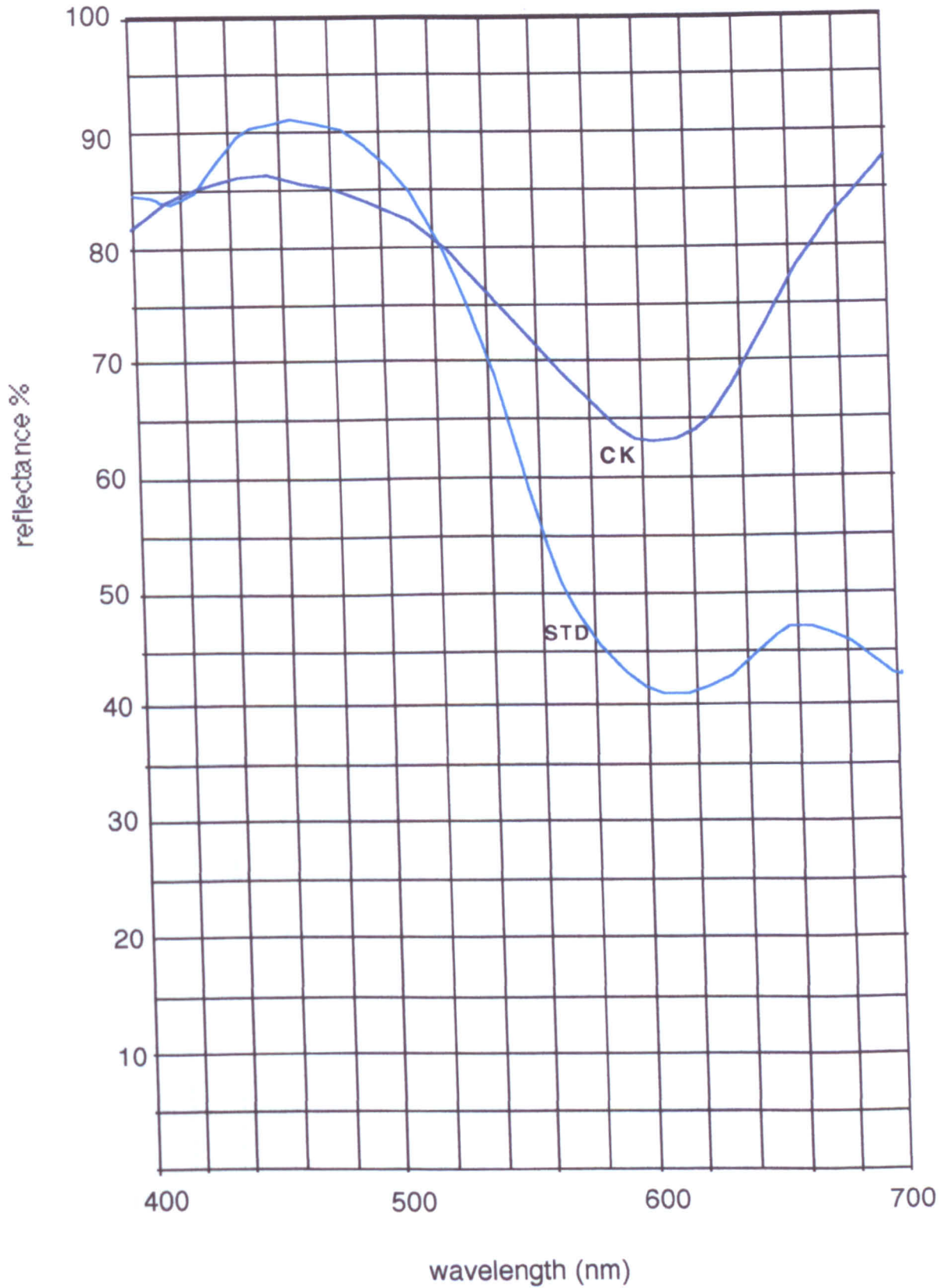
**CD. Sulphonated Indigo Lake.** Prepared as CA and suspended in Rowney System 3 acrylic print medium (12 gm lake/25 ml medium. 1 pull.

## Appendix 4 Spectral Reflection Comparisons



**CG. Indigo Carmine.** A sulphonated synthetic indigo (C I Acid Blue 74) obtained from Sigma Aldrich. Thickened with sodium alginate.

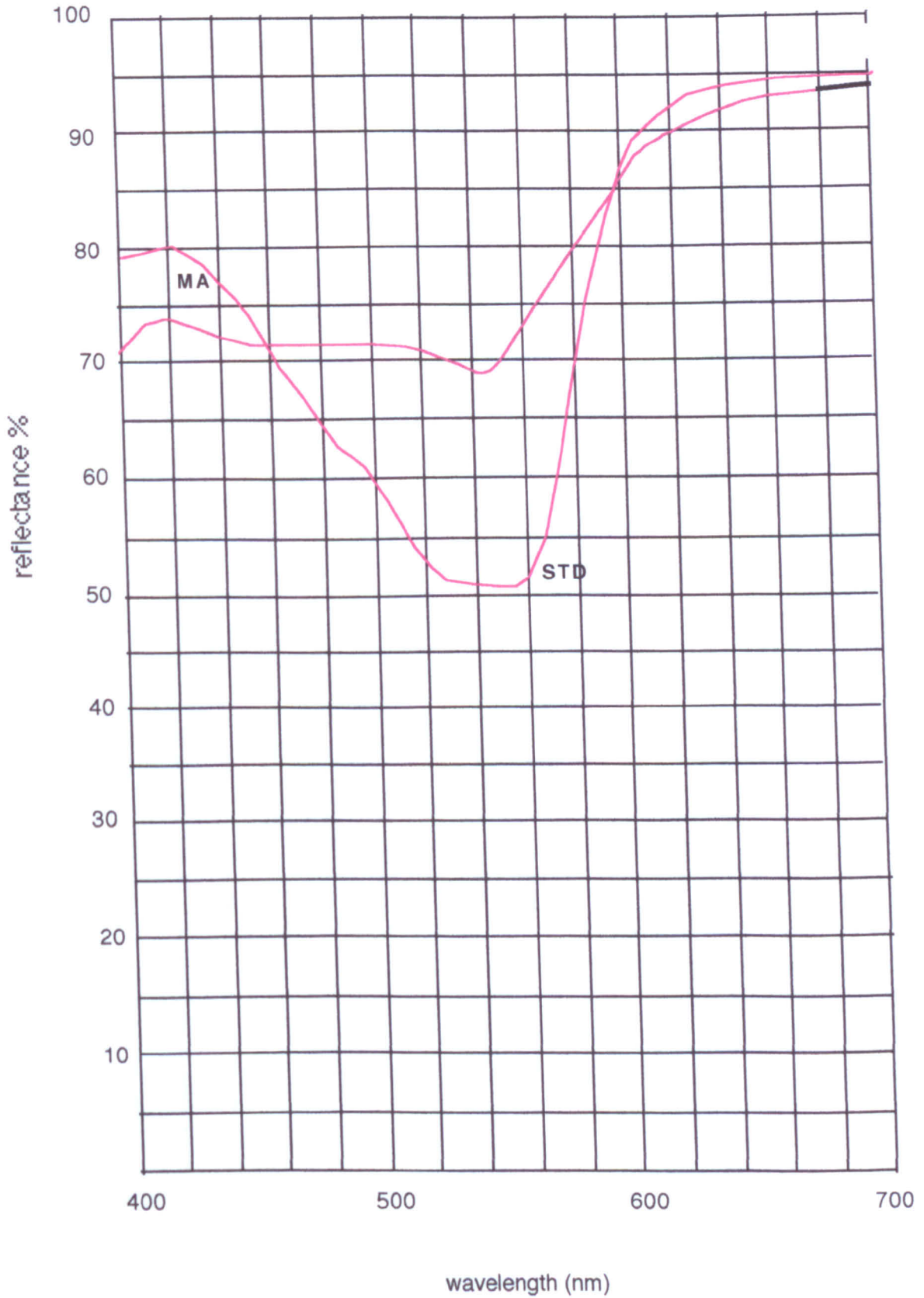
## Appendix 4 Spectral Reflection Comparisons



**CK. Indigo Carmine with 5% weld.** Thickened with sodium alginate.



## Appendix 4 Spectral Reflectance Comparisons



**Brazilwood.** 30 gm brazilwood chips, 2 gm sodium carbonate in 500 ml water heated to 100°C and thickened with sodium alginate.

## Appendix 4 Spectral Reflectance Comparisons



**MB. Pokeweed.** 5 gm frozen berries in 500 ml water heated to 100°C for 60 mins, thickened with sodium alginate

## Appendix 4 Spectral Reflectance Comparisons



**MC. Pokeweed.** 10 gm frozen berries in 250 ml water heated to 70°C for 120 min. Thickened with sodium alginate.

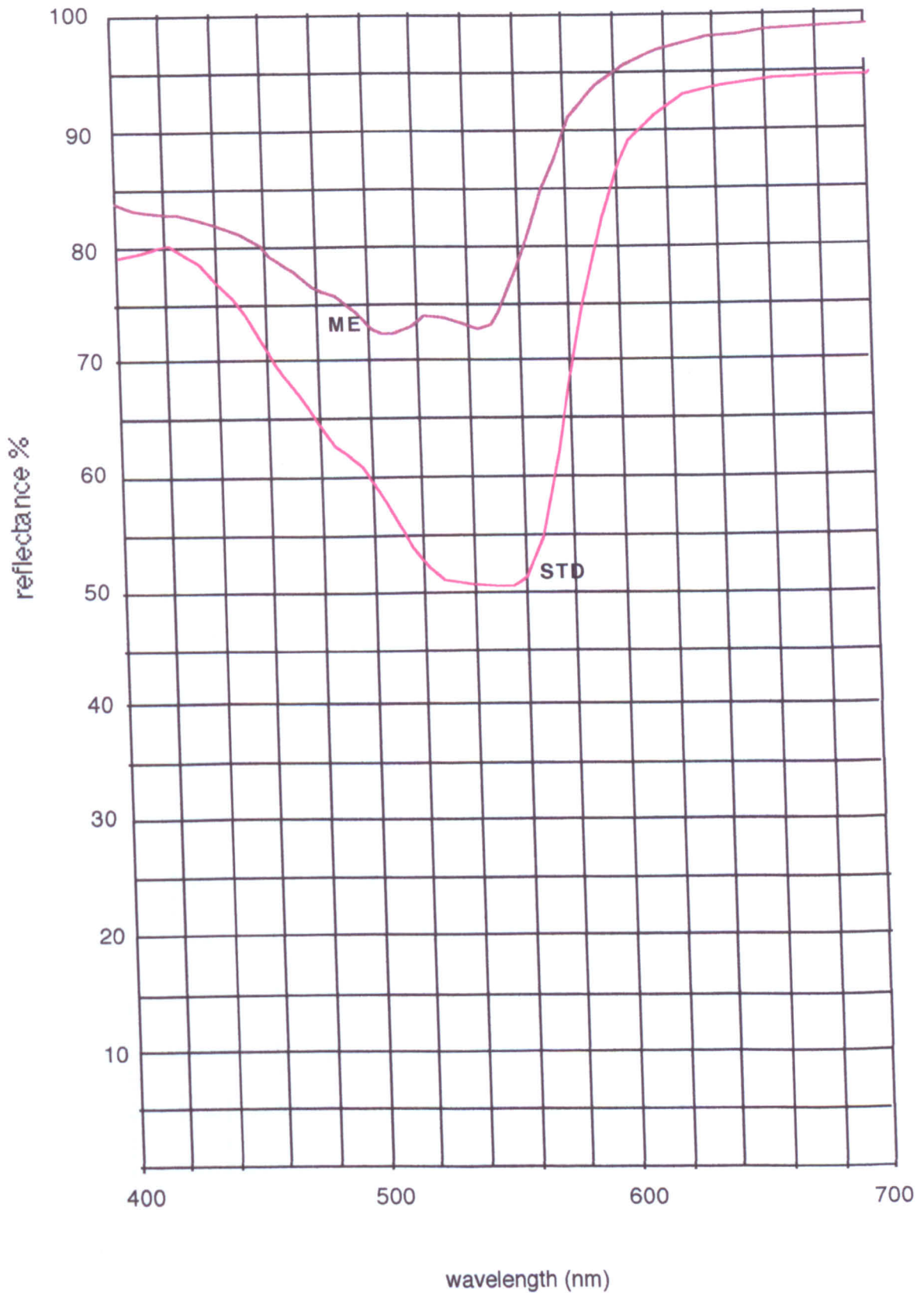
## Appendix 4 Spectral Reflectance Comparisons



**MD. Windsor & Newton Rose Madder.** 2.5 gm lake pigment suspended in 50 ml Rowney System 3 acrylic print medium

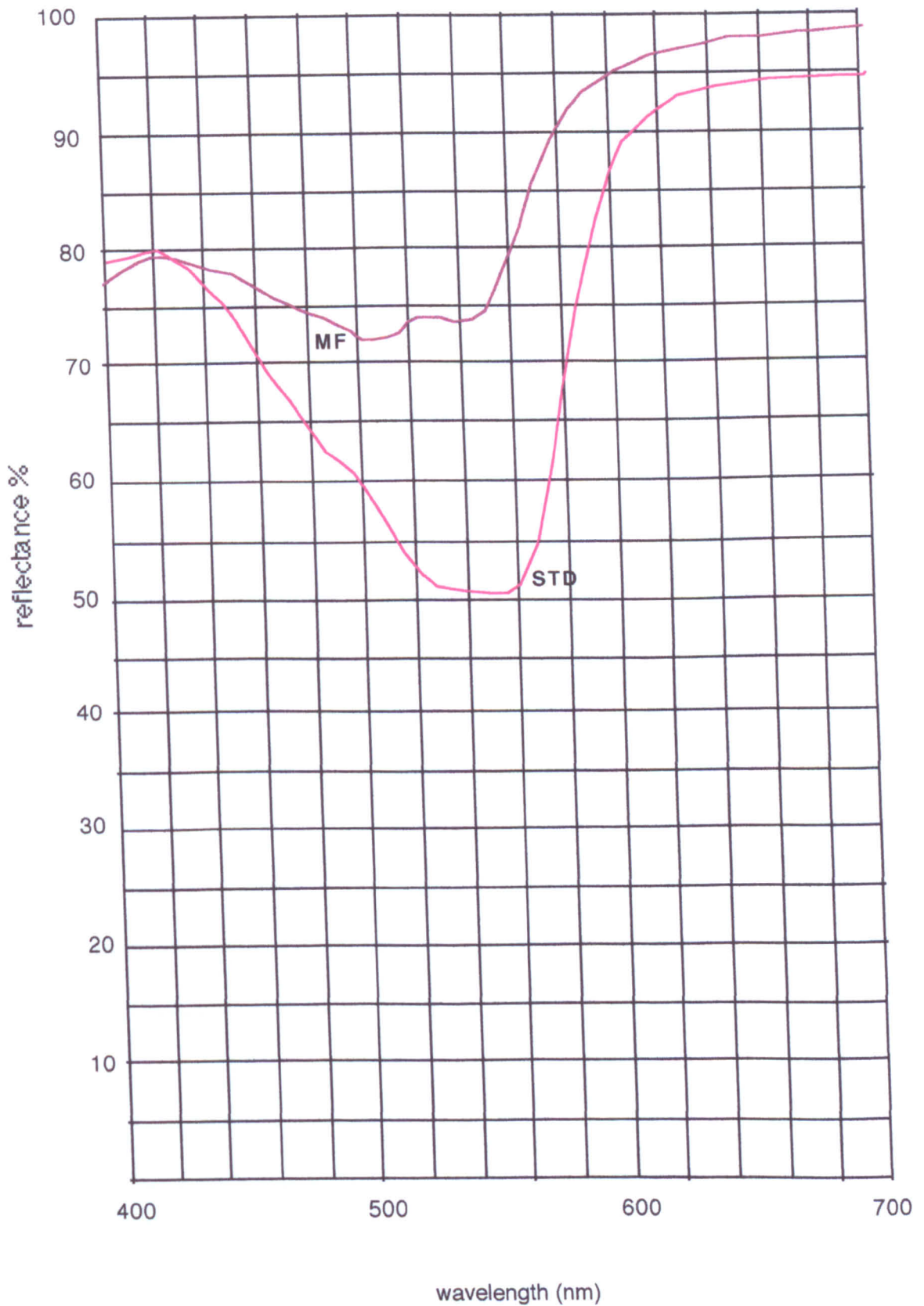


## Appendix 4 Spectral Reflectance Comparisons



**ME. Madder Lake.** 20 gm ground madder root washed in a solution of sodium sulphate, filtered and heated to 100°C in a 10% solution of aluminium sulphate (250 ml) then precipitated onto aluminium hydroxide. Suspended in Rowney System 3 acrylic print medium.

## Appendix 4 Spectral Reflectance Comparisons



**MF. Madder Lake.** As ME but reduced by 50% before precipitation onto aluminium hydroxide. Suspended in Rowney System 3 acrylic print medium.

## Appendix 4 Spectral Reflectance Comparisons



**MG. Brazilwood Lake.** Prepared as MA and precipitated onto aluminium hydroxide. Suspended in Rowney System 3 acrylic print medium.

## Appendix 4 Spectral Reflectance Comparisons



**MH. Windsor & Newton Rose Madder.** 2.5 gm lake pigment suspended in 50 ml Rowney System 3 acrylic print medium.

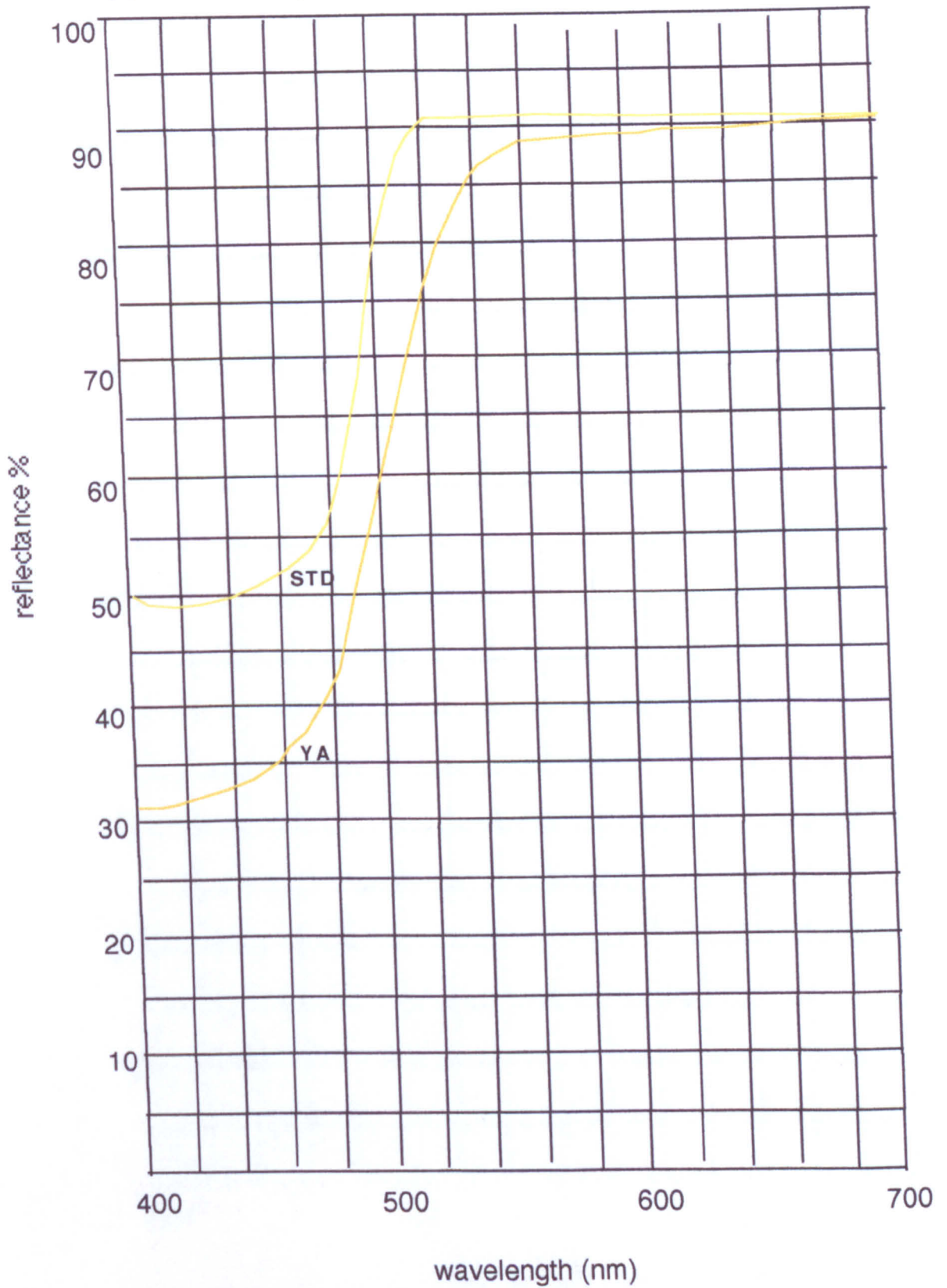


## Appendix 4 Spectral Reflectance Comparisons



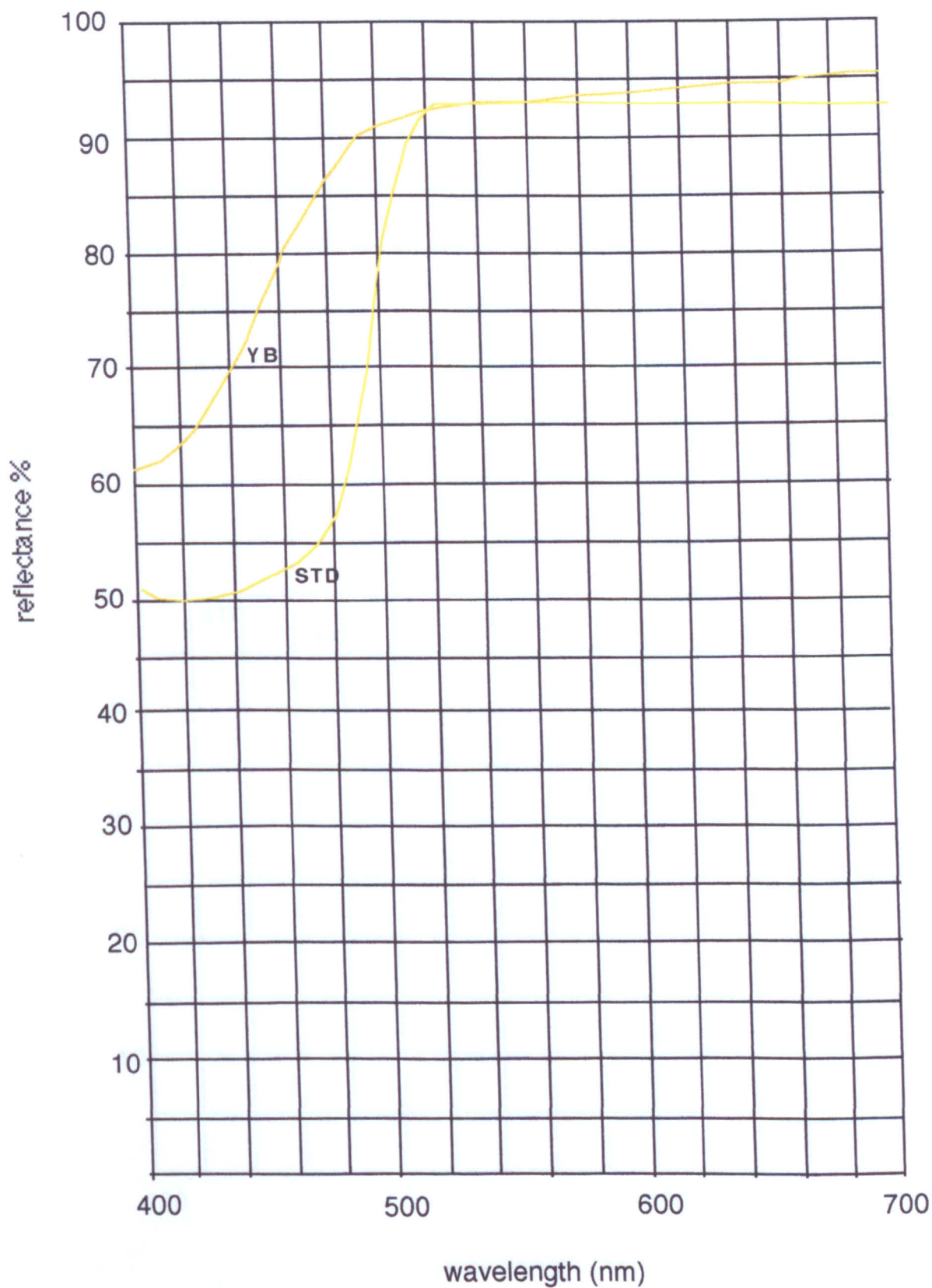
**MI. Cochineal Lake.** 5 gm dried insect, .1 gm aluminium sulphate heated in 125 ml water to 100°C for 10 min and precipitated onto aluminium hydroxide. Suspended in Rowney System 3 acrylic print medium.

## Appendix 4 Spectral Reflectance Comparisons



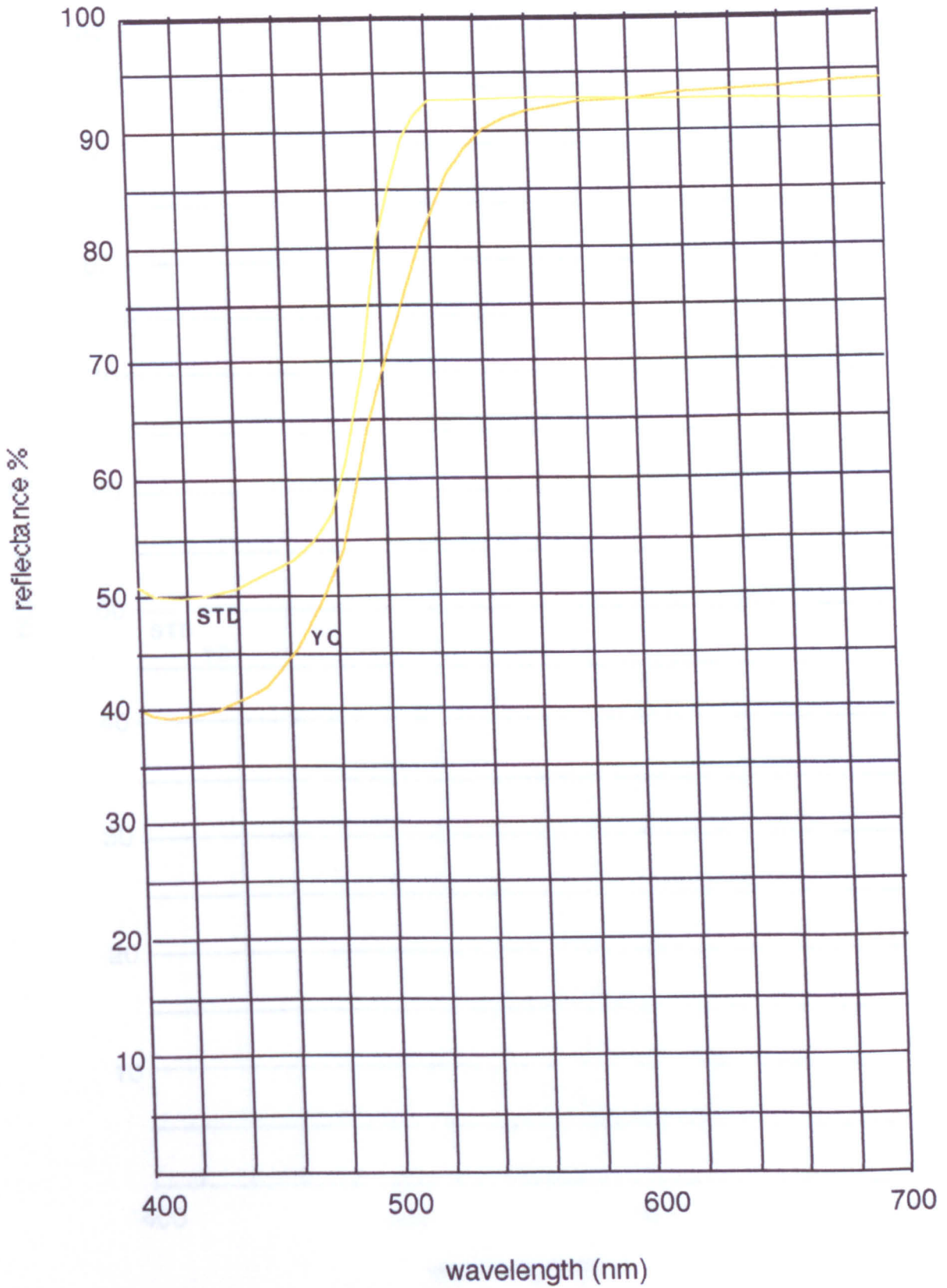
**YA. Persian Berries.** 30 gm persian berries, 1 gm aluminium sulphate, 2 gm sodium carbonate, heated in 500 ml water to 100°C for 75 min. Thickened with sodium alginate.

## Appendix 4 Spectral Reflectance Comparisons



**YB. Weld.** 30 gm dried weld tops, 1 gm aluminium sulphate, 2 gm sodium carbonate, heated in 500 ml water to 100°C for 75 min. Thickened with sodium alginate.

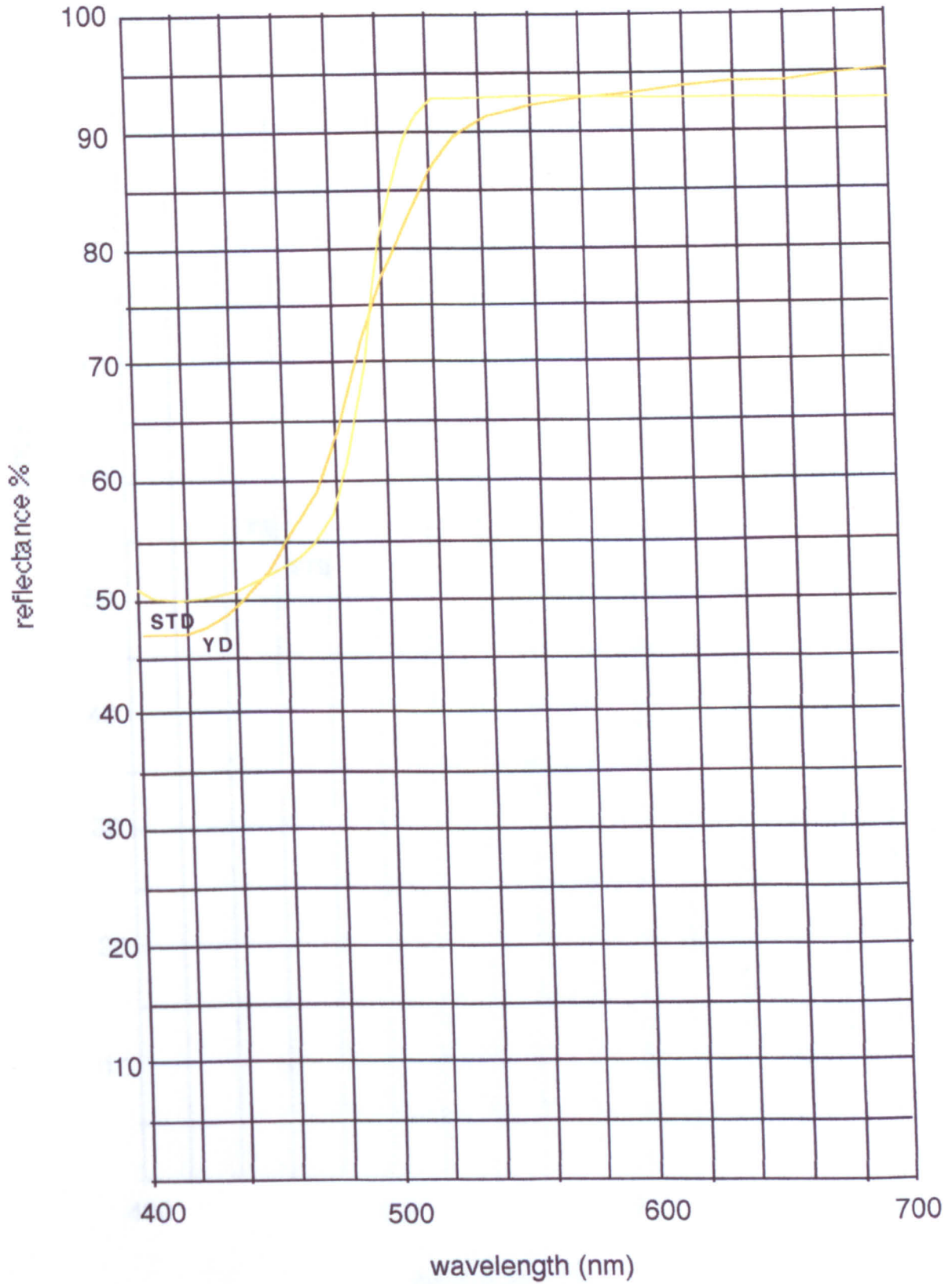
## Appendix 4 Spectral Reflectance Comparisons



**YC. Weld/persian Berry.** 50% weld, 50% persian berry prepared as above. Thickened with sodium alginate.

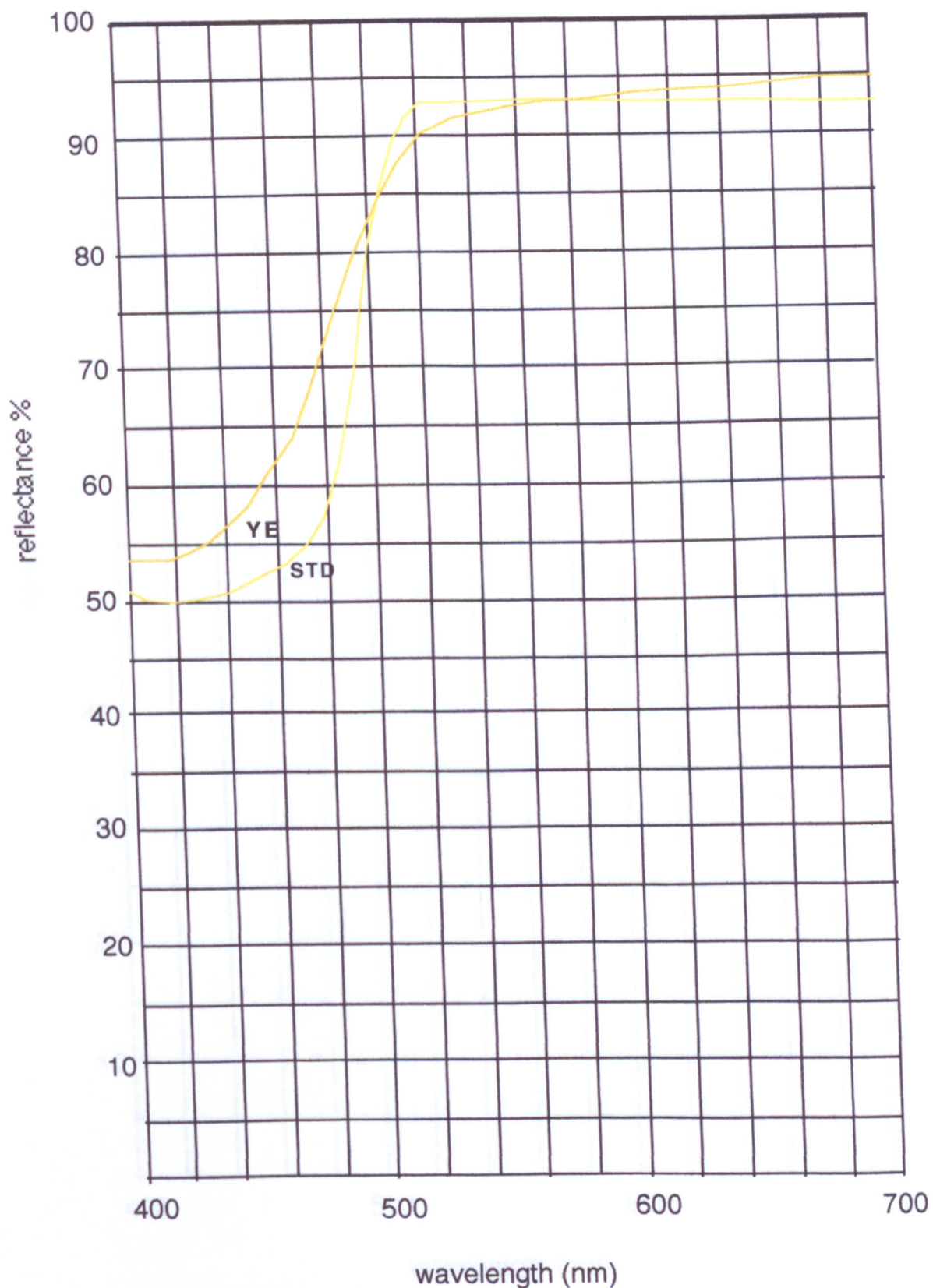


## Appendix 4 Spectral Reflectance Comparisons



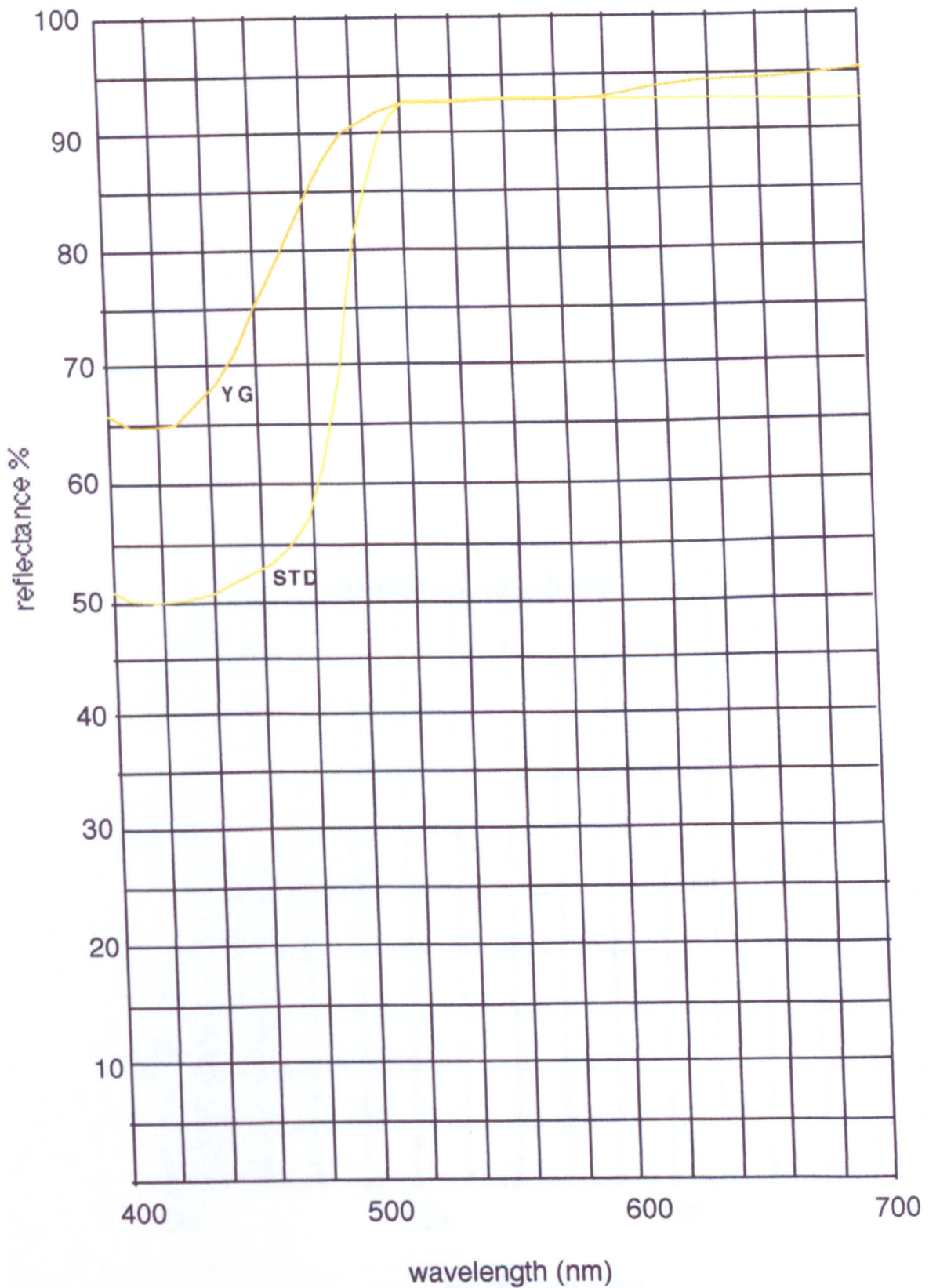
**YD. Weld/Persian Berry.** 75% weld, 25% persian berry. Thickened with sodium alginate

## Appendix 4 Spectral Reflectance Comparisons



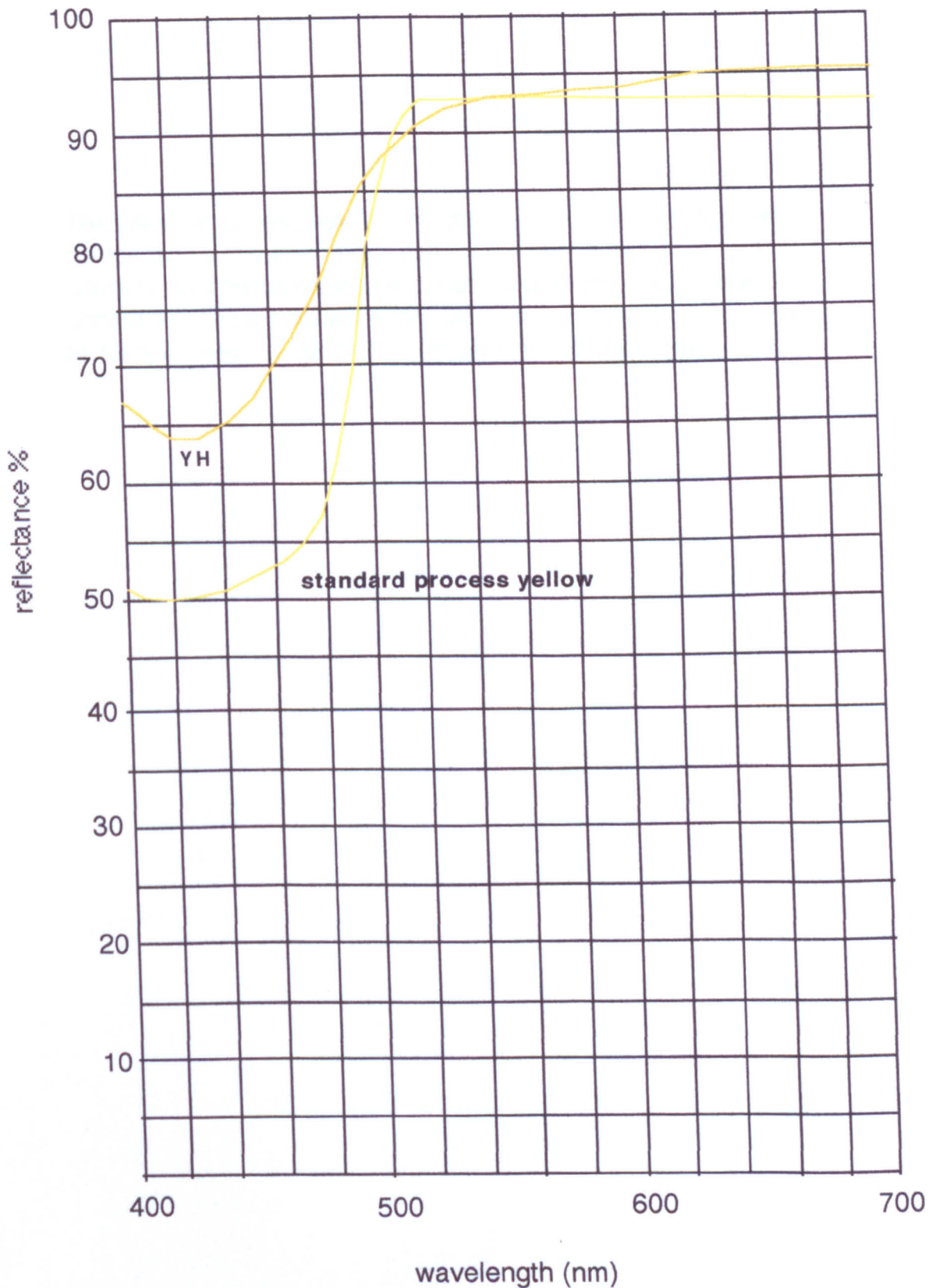
**YE. Weld/Persian Berry.** 87.5% weld, 12.5% persian berries.  
Thickened with sodium alginate.

## Appendix 4 Spectral Reflectance Comparisons



**YG. Weld Lake.** 30 gm dried weld tops, 1 gm aluminium sulphate, 2 gm sodium carbonate, heated in 500 ml water to 100°C for 75 min and precipitated onto calcium carbonate. Suspended in Rowney System 3 acrylic print medium.

## Appendix 4 Spectral Reflectance Comparisons



**YH. Persian Berry Lake.** 30 gm persian berries, 1 gm aluminium sulphate, 2 gm sodium carbonate, heated in 500 ml water to 100°C for 75 min and precipitated onto calcium carbonate. Suspended in Rowney System 3 acrylic



## **Appendix 5 Comparative Lightfastness**

The following plates show the results of a series of light fastness comparison tests which were carried out over a period of 1000 hours of continuous exposure to natural daylight. Each series of printed test strips included a standard process colour sample and all samples were subjected to the same exposure period.

## Appendix 5 Comparative Lightfastness

Daylight exposure (hours)

zero

200

400

600

800

1000



**Natural indigo powder (dark)** suspended in sodium alginate.



**Natural indigo powder (Maya Blue)** suspended in sodium alginate.



**Woad.** 200gm woad leaves heated in 1000 ml water to 100°C, 10 gm sodium carbonate added to the filtered liquor and aerated to form Indigo precipitate. Suspended in sodium alginate.

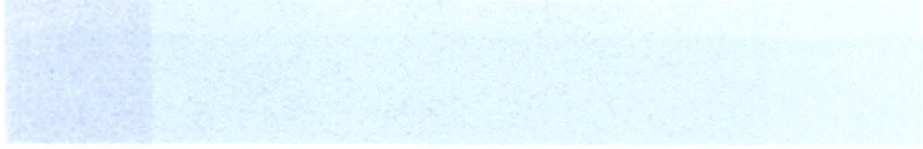


**Standard Process Cyan (Rowney System 3)**

## Appendix 5 Comparative Lightfastness

Daylight exposure (hours)

zero                      200                      400                      600                      800                      1000



**CA.** Sulphonated Indigo 'Lake'. Prepared with 1 part powdered indigo in 5 parts concentrated sulphuric acid, mixed and neutralised with precipitated chalk. The soluble indigo precipitated onto aluminium hydroxide and suspended in sodium alginate and gum arabic.



**CB.** Sulphonated Indigo 'Lake'. Prepared as above and suspended in Rowney System 3 acrylic print medium (6 gm lake/25 ml medium).



**CC.** Sulphonated Indigo 'Lake'. Prepared as CA and suspended in Rowney System 3 acrylic print medium (12 gm lake/25 ml medium. 2 pulls.



**CD.** Sulphonated Indigo 'Lake'. Prepared as CA and suspended in Rowney System 3 acrylic print medium (12 gm lake/25 ml medium. 1 pull.



Standard Process Cyan (Rowney System 3)

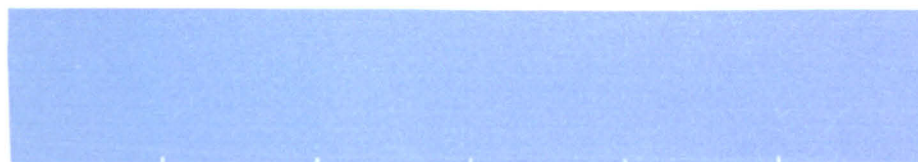
## Appendix 5 Comparative Lightfastness

Daylight exposure (hours)

zero            200            400            600            800            1000



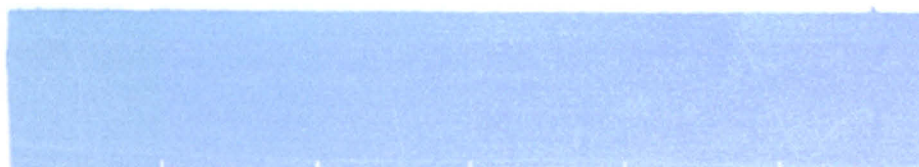
**CG.** Indigo Carmine. A sulphonated synthetic indigo (C I Acid Blue 74) obtained from Sigma Aldrich. Thickened with sodium alginate.



**CH.** As CG + 10% weld



**CI.** As CG + 15% weld



**CJ.** As CG + 20% weld



**CK.** As CG + 5% weld



Standard Process Cyan (Rowney System 3)

## Appendix 5 Comparative Lightfastness

Daylight exposure (hours)

zero                      200                      400                      600                      800                      1000



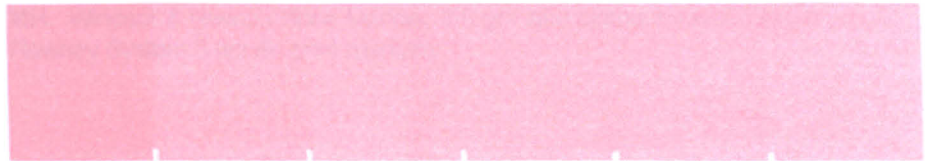
**MA.** Brazilwood. 30 gm brazilwood chips, 2 gm sodium carbonate in 500 ml water heated to 100°C and thickened with sodium alginate.



**MB.** Pokeweed. 5 gm frozen berries in 500 ml water heated to 100°C for 60 mins, thickened with sodium alginate



**MC.** Pokeweed. 10 gm frozen berries in 250 ml water heated to 70°C for 120 min. Thickened with sodium alginate.



**MD.** Windsor & Newton Rose Madder. 2.5 gm lake pigment suspended in 50 ml Rowney System 3 acrylic print medium



Standard Process Magenta (Rowney System 3)



## Appendix 5 Comparative Lightfastness

Daylight exposure (hours)

zero                      200                      400                      600                      800                      1000



**ME.** Madder Lake. 20 gm ground madder root washed in a solution of sodium sulphate, filtered and heated to 100°C in a 10% solution of aluminium sulphate (250 ml) then precipitated onto aluminium hydroxide. Suspended in Rowney System 3 acrylic print medium.



**MG.** Brazilwood Lake. Prepared as MA and precipitated onto aluminium hydroxide. Suspended in Rowney System 3 acrylic print medium.



**MH.** Windsor & Newton Rose Madder. 2.5 gm lake pigment suspended in 50 ml Rowney System 3 acrylic print medium.



**MI.** Cochineal Lake. 5 gm dried insect, .1 gm aluminium sulphate heated in 125 ml water to 100°C for 10 min and precipitated onto aluminium hydroxide. Suspended in Rowney System 3 acrylic print medium.



Standard Process Magenta (Rowney System 3)

## Appendix 5 Comparative Lightfastness

Daylight exposure (hours)

zero                      200                      400                      600                      800                      1000



**YA.** Persian Berries. 30 gm persian berries, 1 gm aluminium sulphate, 2 gm sodium carbonate, heated in 500 ml water to 100°C for 75 min. Thickened with sodium alginate.



**YB.** Weld. 30 gm dried weld tops, 1 gm aluminium sulphate, 2 gm sodium carbonate, heated in 500 ml water to 100°C for 75 min. Thickened with sodium alginate.



**YC.** Weld/persian Berry. 50% weld, 50% persian berry prepared as above. Thickened with sodium alginate.



**YD.** Weld/Persian Berry. 75% weld, 25% persian berry. Thickened with sodium alginate

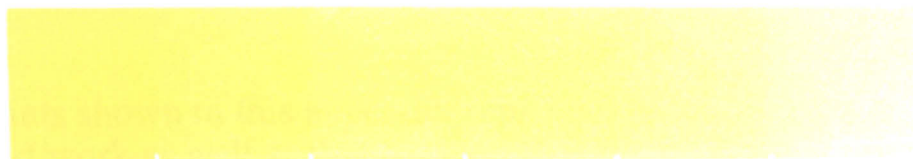


Standard Process Yellow (Rowney System 3)

## Appendix 5 Comparative Lightfastness

Daylight exposure (hours)

zero                      200                      400                      600                      800                      1000



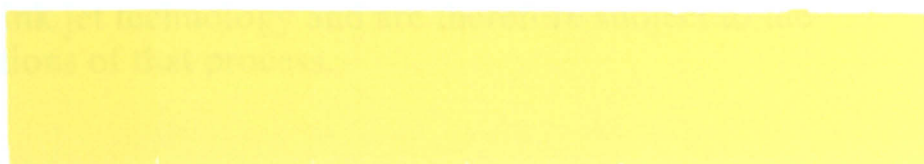
**YE.** Weld/Persian Berry. 87.5% weld, 12.5% persian berries. Thickened with sodium alginate.



**YG.** Weld Lake. 30 gm dried weld tops, 1 gm aluminium sulphate, 2 gm sodium carbonate, heated in 500 ml water to 100°C for 75 min and precipitated onto calcium carbonate. Suspended in Rowney System 3 acrylic print medium.



**YH.** Persian Berry Lake. 30 gm persian berries, 1 gm aluminium sulphate, 2 gm sodium carbonate, heated in 500 ml water to 100°C for 75 min and precipitated onto calcium carbonate. Suspended in Rowney System 3 acrylic print medium.



Standard Process Yellow (Rowney System 3)



## **Appendix 6 Test Prints**

The prints shown in this appendix represent some of my own test work as well as that of several of my students at Middlesex University, the Canadian School for Non-toxic Printmaking and Grafikens Hus in Sweden. Some of the prints were first shown at an exhibition held at the Museum of Garden History in London during the Spring of 1996. They will also form part of an exhibition to be held in the summer of 2000, at Grafikens Hus, a centre for fine printmaking near Stockholm, Sweden. A further series of 35 prints form the visual component of this study one example of which appears in this appendix.

Section 1, contains detailed descriptions of the plant sources, thickeners and other additives used in the four colour separation test prints. Section 2 includes works produced by my students and is captioned with name, title, date, overall size and the plant colours used in their production. Section 3 contains examples of intaglio and lino-cut applications. Unless otherwise stated the thickening agent used was sodium alginate.

It should be noted that the prints are reproduced here using ink jet technology and are therefore subject to the limitations of that process.

## Appendix 6 Section 1 - Four colour Test Prints



Original four colour separation screen print using vegetable sourced CMYK colour, April 1995. The substitute for cyan was extracted from Garden Huckleberry with the addition of sodium carbonate. Magenta from elderberry (*Sambucus nigra*) with potassium aluminium sulphate. Yellow from turmeric (*Curcuma longa*) with no modifiers. Black from oak galls with sodium carbonate and ferrous sulphate. All extracts were thickened with sodium alginate. Screen mesh 90 tpi HD half-tone 65 lpi.

## Appendix 6 Section 1 - Four Colour Test Prints



Four colour separation screen print, September 1995. Cyan from indigo (*indigofera tinctoria*) suspended in sodium alginate. Magenta from elderberry and oregon grape (*mahonia aquifolium*) with potassium aluminium sulphate. Yellow from persian berries with sodium carbonate, black from oak galls combined with sodium carbonate and ferrous sulphate. All extracts thickened with sodium alginate. Screen mesh 90 tpi HD (nylon) half tone 65 lpi.



## Appendix 6 Section 1 Four colour test Prints



Four colour separation screen print, 1996. Cyan from indigo suspended in sodium alginate, magenta from madder, yellow from weld with sodium carbonate and potassium aluminium sulphate, black from logwood. All extracts thickened with sodium alginate. Screen mesh 90 tpi HD (nylon) half tone 65 lpi.

## Appendix 6 Section 1 - Four colour Test Prints



Four colour separation screen print, 1996. Cyan from indigo suspended in sodium alginate, magenta from madder, yellow from weld (*Reseda luteola*) with sodium carbonate and potassium aluminium sulphate, black from logwood (*Haematoxylon campechianum*). All extracts thickened with sodium alginate. Screen mesh 90 tpi HD (nylon) half tone 65 lpi.



## Appendix 6 Section 1 Four colour test Prints



Four colour separation screen print, 1996. Cyan from indigo suspended in sodium alginate, magenta from madder, yellow from weld with sodium carbonate and potassium aluminium sulphate, black from logwood. All extracts thickened with sodium alginate. Screen mesh 90 tpi HD (nylon) half tone 65 lpi.





Appendix 6 Section 1 Four colour test Prints



Four colour separation screen print, 1996. Cyan from sulphonated indigo, magenta from madder lake, yellow from weld with sodium carbonate and potassium aluminium sulphate, black from logwood. All extracts thickened with sodium alginate. Screen mesh 90 tpi HD (nylon) half tone 65 lpi.



Appendix 6 Section 1 Four colour test Prints



Four colour separation screen print 1995. Cyan from sulphonated indigo, magenta from madder lake, yellow from weld, black from logwood. All extracts thickened with sodium alginate. Screen mesh 90 tpi HD (nylon) half tone 65 lpi.

## Appendix 6 Section 1 - Four Colour Test Prints



Four colour separation print, one of a series of 35 which form the visual component of this study, completed in November 1999. Cyan from indigo suspended in sodium alginate with the addition of approximately 5% gum arabic. Magenta from a mixture of madder lake with 5% carmine, a lake of cochineal. Yellow from a combination of 80% weld 20% persian berries with sodium carbonate and potassium aluminium sulphate. Black from a 50/50 mixture of logwood and walnut hulls with sodium carbonate and ferrous sulphate. All extracts thickened with sodium alginate plus approximately 5% gum arabic. Screen mesh 90 tpi HD (nylon) half tone 35 lpi.

## Glossary of Terms and Abbreviations

**Alginates** Sodium salts of alginic acid derived from various species of seaweed, notably *Macrocystis pyrifera* which grows profusely of the pacific coast of the USA and can reach a length of 60 metres.

**Alum** Hydrated aluminium potassium sulphate and related compounds, used as a mordant in textile dyeing, it, along with aluminium hydroxide can also be used to form coloured lakes.

**Annual** A plant that completes its life cycle in a year.

**Artwork** A design prepared for print reproduction.

**Biennial** A plant that completes its life cycle in two years, developing vegetative storage parts in the first year.

**Block printing** Impressions formed on a variety of substrates by designs which have been engraved upon wooden blocks.

**BP No** British Patent Number.

**BS** British Standard, referring here to British Standard 1006, the measure of a pigment or dyes lightfastness.

**Calcerous** In botany, a soil which contains a high proportion of calcium carbonate (chalk).

**Calico** Cotton cloth.

**Chintz** A name given to patterned cotton cloth produced originally on the Coramandel coast of India.

**Chrome** Potassium or Sodium dichromate.

**CI** The standard, internationally recognised catalogue of colourants published by the Society of Dyers and Colourists, known as the Colour Index.

**CIBA Review** A journal published by the Swiss firm now known as Ciba-Geigy containing many articles on plant dyes, pigments and thickeners etc.

**Clogging** In screen printing it refers to the problem of ink drying in the screen mesh, resulting in a partial transference of ink to the substrate. Also called 'filling in.'

**Coal-tar colours** Synthetic dyes derived from coal, first discovered by W.H. Perkin in 1856.

**Colourant** A generic term for any substance which has the power to impart colour to a given substrate.

**Colouring principal** A somewhat archaic term used to define a compound present in animal or vegetable matter, which is responsible for the colours derived from them.

**Copper** Copper or Cupric sulphate, used as a mordant in textile dyeing.

**Copperas** Archaic term for ferrous sulphate.

**Craft dyeing** Any small scale dyeing activity, often using so-called 'natural dyes' as opposed to synthetic, industrial processes.

**Crawling** Movement, or migration of painted or printed colours after application caused by inappropriate formulations, changes in temperature or atmospheric conditions.

**Cyan** A slightly greenish blue typified by the synthetic pigment Phthalocyanine blue, derived from copper.

**Drupe** A fleshy fruit.

**Dye** Any coloured solution, usually aqueous, which can impart its colour to textile fibres.

**Dyestuffs** Dye substances, often used with reference to industrial dye products.

**E** The prefix given in Europe, to permitted food additives, which include colourants, and which is followed by a number, eg. E162 beetroot red.

**Extract** The aqueous dye solution obtained from any plant material.

**Farinaceous starches** Starch obtained from a range of cereal crops.

**Fermentation method** A traditional method used to obtain colour, chiefly from the various species of indigofera.

**Film positives** Photographic or hand rendered positive opaque images on transparent film.

**Flow characteristics** The ability of an ink to flow. (see 'viscosity').

**Fugitive** Colour which does not survive exposure to natural or artificial light without a degree of fading. The term is relative, some colours being more fugitive than others. Very few, if any, dyes and pigments are completely light fast over very long periods of exposure.

**Gall** An excrescence formed on the living plant as a response to the actions of certain insects. Some species of oak and sumac react to the puncturing of young shoots by the female wasp, *cynips sp.* by producing galls which have a high tannin content and which can be used to produce brown and black dyes.

**Gel** Any jelly-like substance.

**Glaucous** Having a green-blue waxy appearance and normally used to describe foliage.

**Globus** Spherical in shape.

**Glucoside** Any one of a number of compounds derived from glucose by hydrolysis. They include most of the plant pigments contained in flowers, a large group of tannins and many drugs.

**Gum** A sticky exudation from many plant species which which harden on exposure to air and can be dissolved in water.

**Herbaceous** Plants which have comparatively soft stems and which generally die down to the ground in autumn.

**Hydrolysis** A chemical reaction in which a compound reacts with water to produce another compound.

**Ink** Any coloured liquid or viscous medium which may be used to make permanent or semi-permanent marks on a given substrate.

**Inflorescence** A cluster or other arrangement of flowers on a plant as opposed to single blooms.

**Iron** In the context of this study iron usually refers to the metallic salt of iron produced when it is brought into contact with hydrogen sulphate (sulphuric acid). It was formerly known as *copperas* (Fe SO<sub>4</sub>)

**JSDC** Journal of the Society of Dyers and Colourists.

**L.** Linnaeus (Carolus Linnaeus 1707-1778) Swedish botanist who established the binomial classification of plant species. The appearance of the letter 'L' after the plant name indicates that Linnaeus was responsible for its generic name (the genus to which it belongs) and specific name (its species within that genus) .

**Lake** A pigment-like material formed by the precipitation of colour upon a near colourless compound eg. Aluminium hydroxide, chalk, etc.

**Lanceolate** Having spear-head shaped leaves.

**Laser scanning** The process by which coloured artwork is separated into the four process colours - yellow, magenta, cyan and black. The scanner optically scans the original and



translates the colour values for each hue and degree of tone into digital format. The apparatus interprets this analysis in terms of the three above hues plus black.

**LF or Light fastness** The ability of a colourant to resist the action of natural or artificial light energy by neither fading, darkening or otherwise changing in character.

**Lithography** A printing process which exploits the lack of affinity between water and oil or grease. It originally involved the use of smooth flat stones (called 'litho stones' and from which the term is derived), latterly zinc, metallised paper or plastic plates have been used.

**Lux hours** The SI unit of illuminance (1 Lux = a luminous flux of 1 Lumen per mtr<sup>2</sup> ) continually transmitted for a given period of time measured in hours.

**Magenta** A colour containing elements of red and violet.

**Mesh** A woven fabric of natural (silk, organdy etc), synthetic (nylon, polyester) or metallic filaments, stretched over a frame and used to support a printing stencil.

**Metallic salts** Salts formed from metals (iron, aluminium, tin etc) by the introduction of acids and a base (alkali).

**Mineral acids** Acids formed from non-organic substances.

**Modifier** Any substance which when added to a dye causes a change in its colour.

**Mordant** A substance which forms a molecular bond, both with the dye and the textile fibre being coloured by it, thereby rendering the colour insoluble in water and thus able to resist the effect of repeated washing. Evidence shows too that these substances can also effect a degree of protection against the influence of light and thus increase light fastness.

**Obovate** Used to describe a leaf that has a stalk at the narrow end, the leaf widening towards the tip.

**ODB** Oxford Dictionary of Botany

**Orchil** A dyestuff obtained from the species of lichen *Roccella tinctoria* and other members of the species. It is also referred to as Archil, Orseille and Orchille.

**Ovary** The collective term for the female reproductive organs of a flower, also termed the gynoecium.

**Oxidation** The addition of oxygen to a compound.

**Panicle** Any compound branched inflorescence.

**Perennial** A plant which survives for more than two seasons and which eventually flowers annually

**Permanent** A term used to describe dyes and pigments which have a high degree of light fastness.

**pH** This refers to the number of hydrogen ions in a substance, in practical terms it provides us with a measure of a given substances acidity or alkalinity - a pH value of below 7 indicates acidity, while a pH value above 7 denotes alkalinity (at 25<sup>o</sup> C.)

**Pigment** Finely divided particles of colour capable of suspension in a liquid medium.

**Pinholing** A fault in printing in which small 'holes' appear in the printed colour, caused by a variety of factors which may the presence of grease on the substrate or inadequate ink formulation.

**Plasticisers** Any high boiling liquids that may be added to an ink formulae in order to improve flexibility and adhesive power.

**Precipitation** The formulation of an insoluble solid by a reaction which occurs in solution.

**Preservatives** An agent, which halts, or slows down the action of microbial or fungal attack.

**Process colours** The four coloured inks used in full colour print reproduction - Yellow, Magenta, Cyan and Black.

**Raceme** A simple unbranched inflorescence in which the flowers are visibly stalked and spirally arranged.

**Resist printing** A method of printing in which a starch, gum, wax or clay is printed onto cloth so as to resist the application of dye, thus leaving the pattern areas reserved in white against a coloured (dyed) background.

**Rhisomatous** A description given to plants which have a horizontal underground creeping stem, which acts as a storage organ.

**Rhizome** see above.

**RHS** Royal Horticultural Society

**Screen** A finely woven mesh stretched over a frame to which stencils may be attached. (see also 'mesh').

**Screen process printing** The modern industrial term given to the method of printing devised in the early part of this century and formerly known as silk screen printing.

**Sessile** A leaf or flower which arises straight from the stem - stalkless.

**Slaked lime** Calcium hydroxide, hydrated lime or hydrate of lime.

**Solvent extraction** The process by which the colour principal is extracted from plant tissue using a solvent eg. water, ethanol, methanol etc.

**Spectral Reflectance** The amount of spectral radiation reflected from the surface of a material, given as a percentage of the incident frequencies of light falling upon it.

**Sp.** Species

**Spike** An unbranched inflorescence in which the individual flowers are stalkless and spirally arranged

**Starch** A polysaccharide (complex carbohydrates) found in all green leaved plants.

**Substrate** The surface upon which ink is printing, eg. paper, cloth, plastic etc.

**Supernatant liquid** The clear liquid above a precipitant which has just settled out.

**Swatch** A small colour sample printed or otherwise applied.

**syn.** Synonym

**Tannin** Tannic acid, a mixture of polyhydroxybenzoic acids found in the barks and other tissues of many trees.

**Thickeners** A range of substances used to increase the viscosity of aqueous colourants, they may include starches, gums and gels.

**Tin** Stannous chloride.

**Tonal range** The continuous change from dark to light seen in any printed reproduction, photograph or original artwork, as distinct from any chromatic (colour) variation.

**Trichromatic** The three 'true' colours used in the four colour process, yellow, magenta and cyan - black being not strictly regarded as as a colour. In practice the term is often used to include black, so that it is in some senses a misnomer.

**Vat method** A method of dyeing textile material or yarn particularly when using indigo, which involves the successive

dipping of the material in large vats of dye so as to achieve deeper shades.

**Viscosity** The ability of an ink to flow, this ability being crucial to successful printing.

**Water-borne coatings** Inks and paints which have water as their solvent base.