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Isolation of colour components from native dye-bearing plants in northeastern India

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Abstract

Recently dyes derived from natural sources have emerged as important alternatives to synthetic dyes. A study was initiated in the year 2000 at the RRL (CSIR), Jorhat to extract dyes from parts of five different plant species indigenous to northeastern India. The colour components responsible for dyeing were isolated and their chemical constituents were established based on chemical and spectroscopic investigations. The principal colour components from the species *Morinda angustifolia* Roxb., *Rubia cordifolia* Linn. and *Tectona grandis* Linn. were found to contain mainly anthraquinone moieties in their molecules. Those from the species *Mimusops elengi* Linn. and *Terminalia arjuna* (Roxb.) Wight & Arn. contained flavonoid moieties in their molecules. The absorption of dye (%) on fibres increased with increasing concentrations of dye in the dye-bath. Maximum absorption of dyes on fibres was obtained at 3% concentration of dyes obtained from *R. cordfolia* (35.350%), *M. angustifolia* (31.580%) and *T. grandis* (25.888%) and at 4% concentration of the dyes from *M. elengi* (31.917%) and *T. arjuna* (12.246%). The K/S values were found to increase with the increase in concentration of mordants. The colour co-ordinates of dyed samples were found to lie in the yellow–red quadrant of the colour space diagram. The dyes obtained from the native plants may be alternative sources to synthetic dyes for the dyeing of natural silk and cotton.

Keywords: Natural dyes; *Rubia cordifolia*; *Morinda angustifolia*; *Tectona grandis*; *Mimusops elengi*; *Terminalia arjuna*; Mulberry silk fibre; Hunter co-ordinates

1. Introduction

Natural dyes/colourants have been used historically throughout the world. The use of natural dyes/colourants has decreased to a large extent due to the advent of synthetic dyes. Recently, dyes derived from natural sources have emerged as important alternatives to synthetic dyes, which have been reported to have carcinogenic effects (Sewekow, 1988). With the worldwide concern over the use of eco-friendly and biodegradable materials, the use of natural dyes has once again gained interest (Eom et al., 2001; Padhy and Rathi, 1990; Garg et al., 1991).

The plant kingdom offers a vast source of natural dyes/colourants which can be obtained from many plant parts e.g., leaves, fruits, seeds, flowers, barks and roots. The sub-Himalayan region of northeastern India has an abundance of plant species with dye-yielding properties. Conventionally, some of the rural folks of the region extract dyes from leaves, roots, flowers or bark of some plant species mostly by boiling, scrapping, powdering and mixing with other materials to get the desired colour.

Though various workers have reported results on the extraction of colour components from different dye-bearing plant species (Onal et al., 1999; Nishida and Kobayashi, 1992a,b; Indrayan and Sharma, 1999), many species remain unexplored. It is critical to explore the extraction of natural dyes from abundantly occurring plant sources.

The objectives of our study were to: (1) extract colour components from selected plant species in the forests of northeastern India, (2) evaluate the chemical constituents of the colour components and (3) examine the dyeing characteristics on mulberry silk and cotton fibres. The plant materials selected for this work are abundant and readily available in different forest areas of northeastern India.

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2. Methods

2.1. Materials

Different parts of five plant species were selected for the present work. These were: (1) stems of *Rubia cordifolia* Linn. (The Indian madder; family: Rubiaceae), a very variable prickly creeper or climber, commonly available throughout India (Chadha, 1972); (2) roots of *Morinda angustifolia* (family: Rubiaceae), a small deciduous shrub, found in the eastern Himalayas and many other states of India (Shastri, 1962; Krishna Rao et al., 1978; Dutta, 1985); (3) leaves of *Tectona grandis* Linn. (family: Verbenaceae), a large deciduous tree indigenous to peninsular India and cultivated in many parts of the country (Chadha, 1976) and (4) barks of *Mimusops elengi* Linn. (family: Sapotaceae) and *Terminalia arjuna* (Roxb.) Wight and Arn. (family: Combretaceae), two large evergreen trees available throughout the country (Chadha, 1976; Dutta, 1985; Shastri, 1962).

Plant parts (5000 g each) were collected from reserve forests near Jorhat (26°4′N latitude and 94°12′E longitude), India, and were harvested manually. The collected plant parts were put into plastic containers and transported to the laboratory. The samples were washed under free flowing water to remove dust and other particles and air-dried. The air-dried samples were stored at ambient temperature in sealed plastic bags.

Bombyx mori silk fibroin produced by following standard de-gumming and spinning techniques (Choudhury, 1992), was collected from a government farm producing such silks. The silk fibroins were processed ready for weaving, so no further purification was done.

The cotton fibre, purchased locally was soaked in distilled water and then scoured in a non-ionic detergent solution containing 2 $\mathrm{g\,l^{-1}}$ each of soap and soda ash at 80 °C for an hour to remove starch and other stiffening agents. The material to liquor (M:L) ratio was maintained at 1:30, after which the fibres were washed with distilled water, dried and ironed. Prior to dyeing or mordanting, the fibre samples were soaked in de-ionized water for half an hour (Mukherjee, 1999).

Mordants such as copper sulphate ($CuSO_4 \cdot 5H_2O$) (LR, CDH), potassium dichromate ($K_2Cr_2O_7$) (LR, CDH), stannous chloride ($SnCl_2 \cdot 2H_2O$) (LR, CDH) and alum [$Al(NH_4)(SO_4)_2 \cdot 12H_2O$] (LR, CDH) were used. Distilled water was used in extraction and for preparation of all chemical solutions. De-ionized water was used for dyeing processes.

2.2. Preparation of samples for extraction of colour components

The air-dried samples containing about 12–15% moisture were ground in a Wiley mill with a 2 mm BIS sieve and stored in plastic bags at room temperature (25–27 °C).

2.3. Extraction techniques adopted for isolation of colour components

Each sample (1000 g) was first extracted with distilled water at 90–95 °C for 3 h in a conical flask (2000 ml). The extract was collected, fresh distilled water was added, and the same procedure was repeated until the colour in the extract was negligible. The total extract was then concentrated under reduced pressure over a water bath to get a solid mass. The solid mass was subjected to extraction with 70:30 (v/v) alcohol:water mixture and filtered. The filtrate was evaporated under reduced pressure to get a concentrated mass of the colour component.

The concentrated colour mass (45 g) obtained from stems of *R. cordifolia* Linn. was subsequently extracted successively with petroleum ether and chloroform and each of the fractions were chromatographed over an open column of silica gel (60–120 mesh) (Itokawa et al., 1989).

Approximately 40 g of the concentrated colour mass from roots of *M. angustifolia* were successively extracted with petroleum ether and benzene. The benzene extract was then passed through an open column of silica gel (60–120 mesh) and eluted with *n*-hexane:ethyl acetate in the ratio 30:1 (v/v) (Bhuyan et al., 2002).

The concentrated mass from leaves of *T. grandis* (about 50 g) was partitioned between chloroform and water. The chloroform extract was dried with anhydrous sodium sulphate, filtered and concentrated. The chloroform extract was then subjected to column chromatography over an open column of silica gel and then eluted with hexane; hexane:ethyl acetate at ratios (v/v) (10:1), (5:1), (7:2), (3:2), (1:1) and ethyl acetate; ethyl acetate:methanol at (10:1), (6:1), (4:1), (1:1) respectively (Aguinaldo et al., 1993).

Approximately 50 g concentrated colour residue obtained from extracted bark of *M. elengi* was partitioned between ether and ethyl acetate. The ether concentrate was chromatographed over silica gel using various ratios of chloroform and methanol and obtained two fractions C-1 and C-2 both of which crystallized as yellow needles. The ethyl acetate concentrate upon column chromatography over silica gel afforded two pure compounds C-3 and C-4 upon repeated crystallization from methanol (Subramanian and Ramachandran Nair, 1973).

Approximately 40 g of the concentrate obtained from the bark of *T. arjuna* was chromatographed on an open column of silica gel in benzene and eluted with benzene, benzene:ethyl acetate, ethyl acetate, ethyl acetate:methanol and methanol. The elution with benzene:ethyl acetate (10:1) afforded fraction F-1 which was further purified by re-column chromatography on silica gel.