



## Impact of natural dye *Nasturtium officinale* R.Br. on non-wood paper properties

Sartaj A Ganie<sup>1\*</sup>, Saakshy Agarwal<sup>2</sup>, MohamadAneesul Mehmood<sup>3</sup>

<sup>1</sup>Division of Environmental Sciences, Shere-Kashmir University of Agricultural Sciences & Technology of Kashmir, Shalimar (India)

<sup>2</sup>Kumarappa National Handmade Paper Institute, Jaipur 302019, Rajasthan (India)

\*Corresponding author

### ABSTRACT

The extracted dye from *Nasturtium officinale* R.Br. has been explored for its application in paper making. The studies have been conducted on aqueous and organic solvent extraction for extraction of natural dye. The extracted natural dye was characterized for the presence of anthocyanin, carotenoid, and chlorophyll content. *Nasturtium officinale* R.Br. recorded highest per cent yield with ethanol and lowest with ethyl acetate solvent. Aqueous extract was applied on 12 % alkaline peroxide pulp of *Daturastramonium*. The strength properties of dyed and undyed has been compared with and it has been found that natural dyed paper has better strength properties. Overall results revealed that *Nasturtium officinale* R.Br. can be used as potent dye yielding plant for pulp and paper industry. The application of natural dyes may offer the great potential for producing different shades to retain its eco-friendly credentials by replacing the synthetic dyes.

**Keywords:** *Daturastramonium*, *Nasturtium Officinaler.Br.* Organic Solvents, Strength Properties.

### I. INTRODUCTION

Today, the protection of environment has become a challenge for the various industries. All over the world, environmental regulations are becoming stricter and forcing the shift of technology towards less or practically non-polluting areas of technological development. The dyestuff industry is under increasing pressure to minimize the damage to the environment, which is caused by their production, application processes and their effluents. Synthetic dyestuffs have also come under severe criticism on the grounds of causing toxic and allergic reactions to humans and also cause carcinogenicity and inhibition of benthic photosynthesis [1].

Environmental problems from the dyeing of paper industry arose after industrialization, when traditional natural dyes were replaced by synthetic dyes. Synthetic dyes are designed to resist chemicals and improve the quality of the product but are persistent in the environment [2]. Most synthetic dyes are carcinogenic, highly toxic in nature and cause allergic dermatitis, skin irritation and mutation to humans [3]. The contamination of water due to synthetic dye molecules causes damage to the environment and has adverse effects on public health [4]. During the dyeing process loss of colourants to the environment can reach 10-

50 percent [5; 6; 7]. Some dyes are highly toxic and mutagenic and also decrease light penetration and photosynthetic activity, causing oxygen deficiency and limiting beneficial uses such as recreation, drinking and irrigation [5; 7].

As a result of the worldwide concern over the carcinogenic effects, toxicity and allergic reactions associated with synthetic dyes interest in the revival of natural dyes is increasing [1]. Moreover, many countries have already imposed stringent environmental standards over synthetic dyes. Nowadays there is a worldwide resurgence of interest in natural dyes, because of increased consumer awareness in terms of minimal environmental and health impact versus synthetic dyes and revival of old practice of coloration with natural dyestuffs [8]; 9] .

In view of the negative impacts of the synthetic dyes and ignorance of natural materials for dyeing, there is dire need to search out new cheaper dye sources from the biodiversity wealth and extraction methods for producing sufficient quantities of viable safe natural pigments and dyes. Therefore, it becomes necessary to develop new techniques of coloration and standardize these processes with the help of modern scientific inputs so that natural dyes can offer themselves as an effective ecofriendly option to their synthetic counterparts. For this *Nasturtium officinale* R.Br. finds an important application in paper industry. Kashmir valley exhibits a great degree of plant diversity. Many workers have contributed to ethno botany and plant diversity studies of the state but dye yielding potential and their application on paper of the flora has not been studied so far. Hence, the study titled “Impact of natural dye *Nasturtium officinale* R.Br. on non-wood paper properties” is proposed.

## **II. MATERIALS AND METHODS**

Plant material for paper making and dyeing was collected from Srinagar and Kulgam districts of Kashmir province of J&K state. The experimental material comprised of 12 % alkaline peroxide pulp *Daturastramonium* L. (Jimson weed) for papermaking, petals of *Nasturtium officinale* R.Br. for dye extraction.

### **2.1 Preparation of dyeing material**

The shade dried fresh plant material of *Nasturtium officinale* R.Br. was washed with water to remove dirt and other adhering materials.

### **2.2 Quantification of pigments**

#### **2.2.1 Total anthocyanin content**

For anthocyanins estimation method of Fuleki and Francis (1968) [10] was followed.

#### **2.2.2 Total chlorophyll and carotenoids**

Chlorophylls and carotenoid content were estimated by Hiscox and Israelstam (1979) [11] and Arnon (1949) [12].

## **2.3 Extraction and dye yield% of selected plant species**

### **2.3.1 Aqueous extraction of dye**

Extraction of dye from selected test species was performed by Soxhlet apparatus using distilled water as solvent. For 100 gram plant material 1 litre of distilled water was used. The material was kept for reflux for about 8 hours at 80-85°C. The liquid extract was evaporated at 65°C in a rotary vacuum evaporator to one fourth of its original volume to get the final extract for dyeing [13].

### **2.3.2 Alcoholic extraction of dye**

Alcoholic extraction of dye from selected test species was performed by Soxhlet apparatus at 80-85°C for 8 hours using different organic solvents ethanol, benzene petroleum ether, methanol ethyl acetate and iso propyl alcohol having different polarities. The material to liquor ratio was kept 10:100 ratio in boiling flask of 500 ml capacity. The reflux was repeated till no trace of color in solvent was seen. The extract was collected by filtering the solution through fritted glass assembly. The liquid extract was evaporated at 65°C in a rotary vacuum evaporator to one fourth of its original volume to get the final extract for dyeing [13].

### **2.4 Dyeing of pulp**

Natural dye extracts were used for the dyeing of pulp using conventional process. The simple impregnation of the pulp in the dye bath is defined as conventional dyeing or stock dyeing. The extracted dye solution (3%) was added to beaten pulp in different dosages and stirred for 5-10 minutes for better mixing and retention of dye on pulp. Dyeing conditions includes 20- 25 °C dyeing temperature (Td) and 15 minutes residence time (Tr). The dyeing was realized using a liquor ratio of 55:1 ml/g. [14].

### **2.5 Production of laboratory sheets and evaluation of paper properties (undyed)**

Laboratory sheets from undyed pulp were formed and were tested by standard TAPPI testing methods.

### **2.5 Production of laboratory sheets and evaluation of paper properties (dyed)**

Laboratory sheets from dyed pulp were formed and were tested by standard TAPPI testing methods.

Laboratory handsheets of 60 GSM (g/m<sup>2</sup>) were formed from undyed and dyed pulp on sheet former by Standard TAPPI testing method T221 cm-99. The hand sheets were conditioned at 27 °C and 65 per cent relative humidity for 24 hours in accordance with standard TAPPI testing method T402 sp-98. After conditioning, the physical strength properties were evaluated as per the Standard testing methods. Tensile index by T494 om-01, tear index by T414 om-98, burst index of handsheets was measured by method T 403 om-97, double fold numbers by T423 cm-98 and brightness was calculated according to the ISO 2470-1

## **III. RESULTS AND DISCUSSION**

The data presented in Table1 revealed that photosynthetic pigments (anthocyanins, carotenoids and chlorophyll) in test species varied significantly. Anthocyanins are water-soluble vacuolar pigments of higher plants abundant in juvenile and senescing plants. Anthocyanins are glycosides of anthocyanidins (also called aglycones) and sugars and span quite a range of colour hues and give rise to the blue, purple, red, orange colour to flowers and fruits of

many plants. The absorption spectra of anthocyanins of plant tissues indicate a strong overlapping of anthocyanins, chlorophyll and carotenoids [15] Lee and Graham, 1986). Anthocyanin synthesis in vegetative organs is induced by different environmental factors [16] Chalker, 1999) and anthocyanins concentration increases during senescence [17]. In the present study total anthocyanin content value in *Nasturtium officinale*R.Br. petals was recorded 71.67 mg/100g. Carotenoids are lipid soluble, yellow, orange and red pigments found in all higher plants. The distinctive yellow colours of light-harvesting carotenoids become more apparent in leaves during autumn when chlorophyll degrades revealing their strong colours. Carotenoids are required for the correct assembly of photosystems [18; 19]. Carotenoids content to the tune of 0.07 mg/g fw was recorded in *Nasturtium officinale*R.Br. petals. Chlorophyll is the green pigment utilized by all higher plants for photosynthesis in response to available sunlight. Chlorophyll is a fat soluble tetrapyrrole pigment, occurring in chloroplasts of green plants, photosynthetic bacteria and algae [20]. It plays a fundamental role in the photosynthesis, being capable of converting sunlight into chemical energy. The most important forms of chlorophyll are chlorophyll a and chlorophyll b, occurring in an approximate ratio of 3:1 [21]. 0.39 mg/g fw chlorophyll content was recorded in *Nasturtium officinale*R.Br. petals. The difference in the pigment (anthocyanin, carotenoids, chlorophyll) contents can be attributed to an integration of genetic and environmental factors which affect the photochemistry, biochemistry, physical diffusion of CO<sub>2</sub> into chloroplasts and activities of non-photosynthetic plant tissues [22]. Photosynthetic efficiency is affected by diverse plant and site factors e.g. properties of leaves, plant architecture, duration of photosynthesis, photosynthesis by organs other than leaves, soil factors such as soil moisture, nutrient availability, and CO<sub>2</sub> released in soil respiration and ambient climatic factors such as air temperature, wind speed, CO<sub>2</sub> concentration, relative humidity, angle of sun, and nature of radiation and thus makes an difference in the composition of the pigments.

**Table 1. Quantification of pigments *Nasturtium officinale*R.Br.**

Parameter	Quantity
Anthocyanins(mg/100 g)	71.67
carotenoids (mg/g fw)	0.07
Chlorophyll (mg/g fw)	0.39

Dye yield is the quantity of dye extract obtained after evaporation of water from the extracted dye solution. The per cent yield of dye extract of the selected plant material varied ranging between 0.72 to 23.39% (Table 2). Petals of *Nasturtium officinale*R.Br. recorded highest (23.39%) per cent yield with metanol and lowest (0.72%) was recorded with ethyl acetate solvent. The low extraction yield of test species with ethyl acetate comply with the studies conducted by [23] Angeliniet al. (1997) and may be probably due to dissolution of only free aglycones[24]. Similar results were observed by the study carried out by [25]. The variation in yield of the dye extract may be attributed to the difference in dissolution of dye and the polarity index of the solvent [26; 27]. The high extraction yield of *Nasturtium officinale*R.Br. with organic polar solvents in comparison to non-polar



or less polar solvents showed that the test species contain high amount of organic polar component [26]. The results are well in agreement with the study conducted by Bushraet al. (2009) [28, 26;27].

Table2. Yield (%) of dye extracts of test species

Solvents	Test species
	<i>Nasturtium officinale</i> R.Br.
Petroleum ether	1.12
Benzene	15.91
Chloroform	1.52
Ethyl acetate	0.72
Ethanol	17.97
Methanol	23.39
Water	1.97

Impact of natural dye had significant influence on tensile strength value of *Nasturtium officinale*R.Br. paper (Table 3). *Daturastramonium*paper exhibited highest (67.33 N.m/g) tensile index with addition of dye and lowest (65.20 N.m/g) was shown without dyeing. This may be attributed to the efficient binding of dye molecules to the cellulosic fibre by forming chemical bridge between dye and fibre through mordant, which gets fixed on the fibre and helps in fixation of natural dyes thus increases the tensile strength properties by addition of dyes and with the addition of mordant [29;26]. *Nasturtium officinale*R.Br.also contains carotenoids, flavonoids and triterpenic alcohols, both in their free and esterified forms [25]. Flavonoids are considered favoured bio compounds as chemotaxonomic markers in plants because they show large structural diversity and are chemically stable [25]. During dyeing of cellulosic fibres, the amount of the dye taken up required agent, to acquire a more fixation (30)

The table reveals that the dyed*Daturastramonium* paper showed the highest (7.51 mN.m<sup>2</sup>/g) tear index while as undyed*Daturastramonium* paper showed the lowest (7.10 mN.m<sup>2</sup>/g) tear index. This is probably due to better fibre bonding and better interaction of *Daturastramonium*fibres with the dye molecule as indicated by higher tear index. The complexation of iron centered ion involved carboxylic group of cellulose fibre of test species with the phenolic group of natural dye, giving more enhancement of tear index properties [31]. Natural dyes enhance the tear index very much than synthetic dyes because of relatively higher possibility of hydrogen bond formation than synthetic dyes [26].

The examination of the data in table 3 indicates that that the dyed *Daturastramonium* paper showed the highest (3.83 kPa.m<sup>2</sup>/g) burst indexwhile as undyedshowed the lowest burst index(3.61 kPa.m<sup>2</sup>/g). This may due to more feasibility of dye molecules with the fibres of test species for hydrogen bonding [29; 26]. In the dyeing of adjective colours, the mordant, having the stronger chemical affinity to the colouring matter, plays the role of the cellulose [32]. The enhancement of burst index with the addition of mordant may be attributed that the



addition of mordant helps in fixation of more and more dyes which increases the bonding between the dye molecule and fibre surface hence increases the burst index properties [27]. The presence of chemical compounds (tannin, flavonoids, resins) in dyeing test species explains their great affinity to the cellulosic fibres. This affinity can be increased by means of mordant which favours the bonding of dye molecule with the cellulosic fibre surfaces [32].

The data indicated that folding endurance (Double fold number) was recorded highest (1091) in dyed *Daturastramonium* dyed paper and lowest (1048) folding endurance (Double fold number) was recorded in undyed paper. The paramount results obtained by *Nasturtium officinale* R.Br. dye may be credited to the efficient binding of dye molecules to the cellulosic fibre by forming chemical bridge between dye and fibre through mordant, which get fixed on the fibre and help in fixation of natural dyes thus increases the folding endurance properties by addition of dyes and with the mordant [29; 26]. The presence of chemical compounds in dyeing test species explains their great affinity to the cellulosic fibres [32].

Application of natural dye with and without mordant showed noteworthy effect on brightness percentage value of *Daturastramonium* dyed paper. The data presented in Table 3 indicated that ISO brightness percentage was highest (59.9%) without the addition of dye and lowest (27.57%) with addition of dye. Addition of dye helps in fixation of more and more dyes which increases the bonding between the dye molecule and fibre surface hence decreases the brightness [27].

**Table 3. Paper properties of undyed and dyed of *Daturastramonium***

Paper sample	Paper properties				
	Tensile Index, N.m/g	Tear Index, mN.m <sup>2</sup> /g	Burst Index, kPa.m <sup>2</sup> /g	Double Fold, No.	ISO brightness, %
<i>Nasturtium officinale</i> R.Br. dyed	67.33	7.51	3.83	1091	27.57
12 % APP undyed	65.20	7.10	3.60	1048	59.9

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## V. CONCLUSION

- Per cent yield of dye extract was recorded highest in *Nasturtium officinale* R.Br. with methanol and lowest with ethyl acetate solvent. Aqueous extract of selected natural dye applied on 12% APP pulp enhanced the strength properties of paper to a greater extent.
- The affinity of the dyes recorded best on *Daturastramonium*. However dyed samples showed better dye quality and strength properties than undyed samples.

Thus, it is concluded from the present study that dye extracted from *Nasturtium officinale* R.Br. can be used for the dyeing of *Daturastramonium* paper adopting stock dyeing method. The selected dye showed major influence on the strength properties of paper can be used for attaining the varieties of beautiful shades.

Utilization of these species for paper production and for paper dyeing shall help in environmental conservation in terms of reducing the stress on forest resources and pollution caused by synthetic dyes.

## REFERENCES

- [1] S. S. Kulkarni, A. V. Gokhale, U. M. Bodake, and G. R. Pathade. Cotton dyeing with natural dye extracted from Pomegranate (*Punicagranatum*) Peel. *Universal Journal of Environmental Research and Technology* 1(2), 2012, 135-139.
- [2] L. Aminoddin, D. and Haji. Functional dyeing of wool with natural dye extracted from *Berberis vulgaris* wood and *Rumexhymenosepolus* root as biomordant. *Iranian Journal of Chemistry and Chemical Engineering*, 29(3), 2010, 55-60.
- [3] A. Srivastava, R. Sinha, and D. Roy, Toxicological effects of malachite green, *Aquata Toxicology*, 66, 2004, 319-329.
- [4] I. Kiran, S. Ilhan, N. Caner, C. F. Iscen, and Z. Yildiz. Biosorption properties of dried *Neurosporacrassa* for the removal of Burazol blue ED dye, *Desalination* 249, 2009, 273-278.
- [5] E. Forgacs, T. Cserhati, and G. Oros. Removal of synthetic dyes from wastewaters: A review. *Environment International* 30(7), 2004, 953- 971.
- [6] M. H. Ben, I. Houas, F. Montassar, K. Ghedira, D. Barillier, R. Mosrati, and Ghedira L. Alteration of in vitro and acute in vivo toxicity of textile dyeing wastewater after chemical and biological remediation, *Environmental Science and Pollution Research International* 19(7), 2012, 2634-2643.
- [7] W. Przystas, G. E. Zabłocka, and S. E. Grabinska, Biological removal of Azo and Triphenyl methane dyes and toxicity of process by products. *Water Air Soil Pollution* 223(4), 2012, 1581-1592.
- [8] L. Moreau, and A. Goossens. Allergic contact dermatitis associated with reactive dyes in a dark garment: a case report. *Contact Dermatitis* 53, 2005 150–154.
- [9] S. D. Gilbert, D. E. Witherspoon, C. W. Berry..Coronal leakage following three obturation techniques. *International Endodontic Journal* 34:2001 293–299.
- [10] T. Fuleki, and F. J. Francis. Analysis of Fruit and Vegetable Products. *Journal of Food Science* 33(1), 1968, 78-83.

- [11] J. D. Hiscox, G. F. Israelstam. Method of extraction of Chlorophyll from leaf tissue without maceration, *Canadian Journal of Botany*, 57, 1779, 1332-1334.
- [12] O. J. Arnon. Copper enzymes in isolated chloroplasts, polyphenol oxidases in *Beta vulgaris*, *Plant Physiology* 24, 1949, 1-14.
- [13] P. Lokesh, and S. M. Kumara. Extraction of natural dyes from *Spathodeacampanulata* and its application on silk fabrics and cotton, *Der ChemicaSinica* 4(1), 2013, 111-115.
- [14] S. G. Papadaki, M. C. Krokida, D. G. Economides, A. G. Vlyssides, and E. G. Koukios, Dyeing capacity of *Eucalyptus globules* L. on chemical pulp: An overview. *Chemical Engineering Transactions* 29, 2012, 7-12.
- [15] D.W. Lee, R. Graham,.Leaf optical properties of rainforest sun and extreme shade plants.*American Journal of Botany* 73: 1986, 1100-1108.
- [16] S.L. Chalker, Environmental significance of anthocyanins in plant stress responses. *Photochemistry and Photobiology* 70: 1999, 1-9.
- [17] K. G. Chang, G. H. Fechner, and H. A. Schroeder.Anthocyanins in autumn leaves of quaking aspen in Colorado, *Forest Science* 35, 1989.229-236.
- [18] B.J. Pogson, H.M. Rissler, H.A. Frank, The roles of carotenoids in photosystem II of higher plants. In: Photosystem II: The Light-Driven Water: PlastoquinoneOxidoReductase; Wydrzynski T, Satoh K (eds). pp: 425-447, 2005, Springer Publishers, Verlag, Netherlands.
- [19] T. Li, J.T. Guthrie,. Colour removal from aqueous solutions of metal-complex azo dyes using bacterial cells of *Shewanella* strain J18 143. *Bioresource Technology* 101, 2009, 4291-4295.
- [20] R.J. Priestley,.Effects of Heating on Foodstuffs.Applied Science Publishers Limited, London, England.417 pp, 1979.
- [21] C. A. Weemaes, V. Ooms, A. M. Van Loey and M. E. Hendrickx.Kinetics of chlorophyll degradation and colour loss in heated broccoli juice.*Journal of Agricultural and Food Chemistry* 47, 1999, 2404-2409.
- [22] I. Zelitch,.Photosynthesis, Photorespiration and Plant Productivity. Academic Press, New York. 347 pp, 1971.
- [23] L. G. Angelini, L. Pistelli, P. Belloni, A. Bertoli, and S. Panconesi.*Rubiatinctorum* a source of natural dyes: agronomic evaluation, quantitative analysis of alizarin and industrial assays, *Industrial Crops and Products* 6, 1997, 303-311.
- [24] M. Tenguria, P. Chand, and R. Upadhyay.Estimation of total polyphenolic content in aqueous and methanolic extracts from the bark of *Acacia nilotica*.*International Journal of Pharmaceutical Science and Research* 3(9), 2012, 3458-3461.
- [25] H.D. Neukiron, M. Ambrosio, J. Dovia, A. Guerriero,.Simultaneous quantitative determination of eight triterpenoid monoesters from flowers of 10 varieties of *Calendula officinalis* L. and characterisation of a new triterpenoid monoester. *Phytochemical Analysis*15,2004, 30-35.
- [26] A. Saakshy, M. Agarwal, R. K. Jain and A. K. Sharma.*Acacia arabica*-A source of natural dye for

handmade paper making. *International Journal of Engineering Research and Technology* 2(12), 2013. 2237-2245.

- [27] S. M. Geelani. Fabric Dye Yielding Potential of Selected Herbaceous and Arboreal Plant Species of the Kashmir Valley. Doctoral thesis, Sher-e-Kashmir University of Agricultural Sciences & Technology of Kashmir, Shalimar, Kashmir, 2014.
- [28] S. Bushra, A. Farooq, A. Muhammad,. Effect of Extraction Solvent/Technique on the antioxidant activity of selected medicinal plant extracts. *Molecules* 14,2009, 2167-2180.
- [29] K. H. Prabhu and A. S. Bhute. Plant based natural dyes and mordants: A review. *Journal of Natural Product and Plant Researches*, 2(6), 2012, 649-664.
- [30] S. Adeel, I.A. Bhatti, A. Kausar, E. Osman, Influence of UV radiations on the extraction and dyeing of cotton fabric with *Curcuma longa* L. *Indian Journal of Fibre and Textile Research* 37, 2012 87–90.
- [31] T. Bechtold, A. M. Ali, and R. A. M. Mussak. Reuse of ash tree (*Fraxinus excelsior* L.) bark as natural dye for textile dyeing: process, conditions and process stability, *Colouration Technology* 123, 2007. 271–279.
- [32] J. Erfurt. A Practical Treatise for the use of Papermakers, Paper stainers, Students and Others. In: The Dyeing of Paper Pulp. pp: 106-108. 1901 Scott, Greenwood and Company, London