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Extraction, Application, Chemical Characterization of Natural Dye from *Calendula offcinalis* L.

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ABSTRACT

In the present study Callandulaoffcinalisan ornamental planthas been explored for its application in paper making. The study has been carried out on aqueous and organic solvent for extraction of natural dye. Natural dye was characterized for the presence of anthocyanin, carotenoid, chlorophyll content. Calandullaoffcinalis L. recorded highest per cent yield with ethanol and lowest with ethyl acetate solvent. Aqueous extract, as eco-friendly source was applied on 12 % alkaline peroxide pulp of Daturastramonium. The characteristic strength properties of both dyed and undyed paper was determined and it was found that natural dyed paper showed better strength properties. The overall outcome of the results revealed that Calendula officinalis L. can be used as effectivesource of dye yielding plant for pulp and paper industry. The application of natural dyemight offer the prodigious potential for producing diverse shades to retain eco-friendly credentials by replacing the synthetic dyes, which are mainly carcinogenic in nature.

Keywords*: *Callandulaoffcinalis, Daturastramonium, Organic Solvents, Strength Properties.

I INTRODUCTION

Now a day's environment is deteriorating at an alarming rate especially by industrial pollution. Dyeing of paper industry has also in race of deterioration of environment, 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 obstinate in the environment [1]. Most synthetic dyes are carcinogenic, highly toxic in nature and cause allergic dermatitis, skin irritation and mutation to humans [2]. In recent years, environmental contamination by synthetic dyes is a serious problem due to their negative ecotoxicological effects [3]. The contamination of water due to synthetic dye molecules causes damage to the environment and has adverse effects on public health [4]. The discharge of highly coloured synthetic dye effluents into inland and coastal waters is an environmental problem of growing concern [5]. During the dyeing process loss of colourants to the environment can reach 10-50 percent [6,7]. Some dyes are highly toxic and mutagenic [8].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 [9].

The word 'natural dye' covers all the dyes derived from the natural sources like plants, animal and minerals. Natural dyes are mostly known to be eco-friendly, biodegradable, low toxic and less allergenic as compared to synthetic dyes [10]. The natural dyes are clinically safer than their synthetic analogues, in handling and use because of non-carcinogenic and biodegradable nature [1]. Natural colourants are simple and harmonized with nature, obtained from renewable sources and also their preparation involves a minimum possibility of chemical reactions.

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. The natural dyes can offer themselves as an effective ecofriendly option to their synthetic counter parts. For this *Calendula officinalis* L. finds an important application in paper industry.

Calendula officinalis is one of the most known and used medicinal plants, that is widely cultivated both for obtaining extracts used in phytotherapy and ornamental purposes. *Calendula officinalis* (pot marigold, common marigold, garden marigold, English marigold, or Scottish marigold) is a plant in the genus Calendula of the family Asteraceae. 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 "Extraction, Application, Chemical Characterization of Natural Dye from *Calendula offcinalis* L." 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 *Calendula offcinalis* L. (Marigold), for dye extraction.

Preparation of dyeing material

The shade dried fresh plant material of *Calandullaoffcinalis* L. (Marigold), was washed with water to remove dirt and other adhering materials.

Quantification of pigments

Total anthocyanin content

For anthocyanins estimation method of Fuleki and Francis (1968) [11] was followed.

2.2.2 Total chlorophyll and carotenoids

Chlorophylls and carotenoid content were estimated by Hiscox and Israelstam (1979) and Arnon (1949) [12,13].

2.3 Extraction and dye yield% of selected plant species

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^{o}C. The liquid extract was evaporated at 65^{o}C in a rotary vacuum evaporator to one fourth of its original volume to get the final extract for dyeing [14].

2.4 Yield of dye

From the extracted dye the per cent yield of the natural colourant was calculated after the liquid extract was evaporated at 65^{o}C in a rotary vacuum evaporator. The dried extract was collected, stored in a desiccator and used for estimation of the per cent dye yield by applying the following formula [15].

2.5 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 ^{o}C dyeing temperature (Td) and 15 minutes residence time (Tr). The dyeing was realized using a liquor ratio of 55:1 ml/g[16].

2.6 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.

2.7 Production of laboratory sheets and evaluation of paper properties (dyed with and without mordant)

Laboratory sheets from dyed pulp without mordant were formed and were tested by standard TAPPI testing methods.

Laboratory handsheets of 60 GSM (g/m^2) were formed from undyed and dyed pulp on sheet former by Standard TAPPI testing method T221 cm-99. The handsheets were conditioned at 27 ºC 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

Table1 revealed that photosynthetic pigments (anthocyanins, carotenoids and chlorophyll) in test species varied significantly. Anthocyanin synthesis in vegetative organs is induced by different environmental factors and

anthocyanins concentration increases during senescence [17]. In the present study total anthocyanin content value in *Calendula offcinalis* petals was recorded 10.29 mg/100g. The distinctive yellow colours of light-harvesting carotenoids become more apparent in leaves during autumn when chlorophyll degrades revealing their strong colours. Carotenoids content to the tune of 1.40 mg/g fw was recorded in *Calendula offcinalis* petals.Chlorophyll is the green pigment utilized by all higher plants for photosynthesis in response to available sunlight. The most important forms of chlorophyll are chlorophyll a and chlorophyll b, occurring in an approximate ratio of 3:1 [18]. 4.16 mg/g fw chlorophyll content was recorded in *Calendula offcinalis* 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_2 into chloroplasts and activities of non-photosynthetic plant tissues [15].

Table1. Quantification of pigments *Calendula offcinalis*

Parameter	Quantity
Anthocyanins(mg/100 g)	10.29
carotenoids (mg/g fw)	1.40
Chlorophyll (mg/g fw)	4.16

The per cent yield of dye extract of the selected plant material varied ranging between 2.1 to 27.5% (Table-2). Petals of *Calendula offcinalis* recorded highest (27.5%) per cent yield with ethanol and lowest (2.1%) was recorded with ethyl acetate solvent. The low extraction yield of test species with ethyl acetate comply with the studies conducted by[19] and may be probably due to dissolution of only free aglycones[20].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 [21, 22]. The results are well in agreement with the study conducted by [21, 22].

Table 2. Yield (%) of dye extracts of test species

Solvents	Test species
	Calendula offcinalis
Petroleum ether	4.9
Benzene	23.3
Chloroform	2.9
Ethyl acetate	2.1
Ethanol	27.5
Methanol	26.6
Water	3.4

Application of natural dye had significant influence on tensile strength value of *Daturastramonium* paper (Table 3). *Daturastramonium*paper exhibited highest (78.2 N.m/g) tensile index with addition of dye and lowest (65.2 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, 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 [23].

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The table 3 also revealed that the dyed*Daturastramonium* paper showed the highest (8.6 $mN.m^2/g$) tear index while as undyed*Daturastramonium* paper showed the lowest (6.1 $mN.m^2/g$) 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 [24]. Natural dyes enhance the tear index very much than synthetic dyes because of relatively higher possibility of hydrogen bond formation than synthetic dyes [21,22].

The examination of the data in table 3 indicates that that the dyed *Daturastramonium* paper showed the highest (4.5 $kPa.m^2/g$) burst indexwhile as undyedshowed the lowest burst index(3.6 $kPa.m^2/g$). This may due to more feasibility of *Calendula officinalis*dye molecules with the fibres of test species for hydrogen bonding [21,22]. In the dyeing of adjective colours, the mordant, having the stronger chemical affinity to the colouring matter, plays the role of the cellulose [25].

The data indicated that folding endurance (Double fold number) was recorded highest (1225) in dyed *Daturastramonium* dyed paper and lowest (1048) folding endurance (Double fold number) was recorded in undyedpaper. The paramount results obtained by *Calendula officinalis* 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 [21,22]. Application of natural dye with 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 (17.1%) with addition of dye. Addition of dye increases the bonding between the dye molecule and fibre surface hence decreases the brightness[22].

Table 3. Paperproperties of undyed and dyed of *Daturastromonium*

	Paper properties				
Paper sample	Tensile Index, N.m/g	Tear Index, $mN.m^2/g$	Burst Index, $kPa.m^2/g$	Double Fold, No.	ISO brightness, %
Calendula dyed	75.1	8.4	4.4	1192	20.1
12 % APP undyed	65.2	7.1	3.6	1048	59.9

IV.CONCLUSIONS

Per cent yield of dye extract was recorded highest in *Calendula offcinalis* with ethanol and lowest with ethyl acetate solvent. Aqueous extract of selected natural dye on 12% APP pulp enhanced the strength properties of paper to a greater extent.

The affinity of the dyes recorded best on *Daturastramonium* and showed better dye quality and strength properties than the undyed samples.

Thus, it is concluded from the present study that dye extracted from Calendula officinalis 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.

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