



Production of a concentrated natural dye from Canadian Goldenrod (*Solidago canadensis*) extracts

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ARTICLE INFO

Article history:

Received 23 July 2011

Received in revised form

10 September 2011

Accepted 9 October 2011

Available online 19 October 2011

Keywords:

Natural dye

Textile dyeing

Total phenolics

Precipitate

Canadian Goldenrod

Solidago canadensis

ABSTRACT

The dyestuff content in plant sources is rather low, usually in the order of a few % of the mass of dry plant material. Introduction of plant dyes into technical scale textile dyeing operations thus requires handling, extraction and disposal of huge amounts of plant material.

The precipitation of a solid, dyestuff-containing residue by addition of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ to the aqueous plant extract yields a highly concentrated plant dye. In this work Canadian Goldenrod (*Solidago canadensis*) was used as representative case to study production of a concentrated solid plant dye. An iron content of 5% w/w of the dry precipitate was analysed by photometry (1,10-Phenanthrolinechloride). The content of total phenolics (TPH) calculated as gallic acid monohydrate equivalents according the Folin-Ciocalteu method, was determined with 45% w/w.

The dyestuff precipitate was tested in standard dyeing experiments. Shade and colour depth were found comparable to dyeings obtained with direct use of plant extracts. Use of a concentrated natural dye product offers new approaches with regard to standardisation of dyestuff quality, handling and applicable dyeing techniques.

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1. Introduction

The invention of synthetic colourants at the end of the 19th century triggered a marked decrease in the wide and exclusive use of natural colourants for colouration of materials. Synthetic dyes were judged “better” in any aspect of use, such as brilliance and range of colour, durability and costs. As a result only limited research activities were executed during the 20th century, a considerable part of them located in the field of conservation, restoration and also analysing traditional dyeing procedures as part of cultural history [1–3].

During the last few decades growing interest in the use of natural colourants can be recognised, both in public awareness and scientific activity [4–7]. Nowadays more “soft” aspects about possible use of natural colourants are brought into the discussion. New arguments consider terms like increased sustainability, renewable resources, environmentally friendly processing, reduced pollution, green chemistry [8].

In many cases traditional procedures of natural dyeing cannot be transferred into modern dyehouses, and complete redesign is essential. The historical procedures often exhibit low productivity due to a high number of consecutive steps, use of hazardous metal mordants e.g. copper, tin salts, and also poor efficiency with regard to water and energy consumption.

As a result a number of aspects has to be considered when the re-introduction of natural colourants into technical dyeing processes is attempted [8,9]. Important aspects which also define experimental conditions used for this study are:

- The dyestuff content in the plant material is low, thus an extraction has to be performed with water only. Otherwise huge amounts of chemically contaminated extracted plant material will be released from the extraction step, which is unacceptable with regard to both costs and overall ecological profile of the process.
- Huge amounts of plant material have to be handled during harvesting, storage and extraction.
- Standardisation of the dyestuff is required to minimise variability in the dyeing result due to quality differences of plant material.
- The application of natural colourants must be possible on the technical equipment available in modern textile dyehouse, without big additional investment.

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In any case the mass and energy balances of a proposed process have to be considered with care and must be compared to the present state of the art processes based on synthetic dyes [9].

An extraction of plant material at the site of the textile dyehouse could be accepted during the initial phase of a scale-up of natural dyeing into commercial production, on the long term the success of natural colourants for dyeing processes will depend on the availability of concentrated dyestuff formulations [8].

While the costs of plant material available from direct farming can be reduced by use of by-products from food processing and timber industry, the low dyestuff content present in plant sources causes the central problem for a re-introduction of plant material into commercial textile dyeing processes. As an estimate, for dyeing of 1 kg textile material a dyer will have to process almost the same amount of plant material during dyestuff extraction. From cooperation projects with technical dyehouses this situation was identified as a key problem to be solved before a wider application could be considered [10,11].

A concept to overcome this existing technical barrier is the production of a solid natural colourant by precipitation of colourants from the plant material extracts.

A general principle to obtain a solid dyestuff formulation bases on the insolubility of many metal complexes of natural dyes [12–14]. This reaction already is applied during the mordanting procedures with use of metal salts [15–17]. Due to existing waste water limits and requirements for eco-textiles nowadays only iron and aluminium salts mordants are acceptable for natural dyeing [8]. In previous studies a number of plant sources were identified, which offer potential for future use as natural colourants for textiles, among them Canadian Goldenrod, high tannin content barks, and by-products from food industry e.g. onion peel [1,15]. Among the main criteria for selection of a plant source was availability of the material and fastness properties of the dyed textiles.

In this paper we report a model study to formulate a solid dyestuff product from aqueous extracts of dried Canadian Goldenrod plant material. The dyestuff containing product was precipitated as insoluble metal complex from the aqueous dyestuff solution [18,19]. Basic analytical parameters of the solid plant dye e.g. content of total phenolics, metal content have been analysed. Dyeing behaviour and colour strength have been tested in dyeing experiments and results were compared to dyeings obtained with direct use of aqueous plant material extracts.

2. Experimental

2.1. Chemicals and reagents

Analytical grade chemicals were used for the phenol analysis (Na_2CO_3 , Merck, Darmstadt; gallic acid monohydrate, Riedel-de-Haen, Seelze, Germany; Folin-Ciocalteu reagent, Sigma–Aldrich Chemie, Steinheim, Germany).

For the dyestuff precipitation and dyeing processes $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ (technical grade >96% purity, Riedel-de-Haen), technical grade oxalic acid $\text{H}_2\text{C}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$ (Riedel-de-Haen), $(\text{NH}_4)_2\text{Fe}(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$ and sodium acetate (98.5% purity, Roth, Karlsruhe, Germany) were used.

2.2. Plant material – extraction of dye

Canadian Goldenrod was collected from wild growth in the western region of Austria. The plant was used as a whole including buds, part of stem and upper part of leaves. The material was dried at room temperature and stored in dark.

A weighted amount of dry plant material e.g. 80 g was extracted with distilled water (800 ml) in a beaker. In the standardised

procedure the ratio mass of plant material:volume of liquid (liquor ratio) was kept constant with 1:10. The extraction was performed for approx. 60 min. at 95 °C in an open stainless steel beaker. Manual stirring was sufficient to distribute the plant material in the liquid during the extraction period. Volume loss due to evaporation was compensated by addition of water at the end of the extraction period to obtain the initial volume.

The increase in absorbance of the extracts was analysed as function of extraction time in the wavelength interval of 400–700 nm. For this measurement the extracts were filtered through a paper filter and the filtrate was diluted with 10 times the volume of distilled water. Absorbance was measured using a 10 mm cuvette and a diode-array spectrophotometer (Zeiss CLH 500/MCS521 UV–vis, Carl Zeiss, Jena, Germany).

In the extracts also the content of total phenolics was determined as function of time.

A potentiometer equipped with a glass electrode was used for pH measurement (MP225 pH Meter, Zeller, Hohenems, Austria).

2.3. Precipitation of dyestuff

The extract was filtered through a bleached cotton cloth to remove plant particles. pH was adjusted to pH 7.0 by addition of 2.6% (w/w) ammonia solution. Precipitation was initiated by addition of a 0.18 M (50 g L^{-1}) $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ solution. To a volume of 200 ml extract, a volume of 5–40 ml of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ solution was added at room temperature. After 2 h of gentle stirring, pH of the solution was measured and the precipitate was allowed to settle overnight. Filter paper was used for filtration of the solids (Whatman, 595/1/2, 110 mm diameter). The filtered dye containing product then was dried in a laboratory oven at 60 °C for at least 6 h, and weighted.

2.4. Determination of total phenolics (TPH)

Total soluble phenolics (TPH) in the extract were determined with Folin-Ciocalteu reagent according to the method of Slinkard and Singleton using gallic acid monohydrate as standard [20–22]. The extracts were diluted with distilled water to adjust extinction within the range of the calibration curve. To a volume of 0.2 ml of the diluted extract deionised water (1.4 ml), Folin-Ciocalteu reagent (0.1 ml) and saturated Na_2CO_3 (0.3 ml) solution were added. After a reaction time of 30 min at a temperature of 40 °C, the absorbance was measured at 765 nm (Hitachi U-2000 double-beam spectrophotometer). Results were expressed as both mg L^{-1} TPH in extract or g kg^{-1} TPH dry plant material, calculated as gallic acid monohydrate equivalents [20–22].

For analysis of the TPH content in the solid dyestuff 0.1 g of the dried filter cake was dissolved in a solution of 1.6 mmol (0.2 g) of oxalic acid dihydrate in 10 ml water. Before TPH analysis, this solution then was diluted further by a factor of 10–100.

2.5. Iron content in precipitate

Formation of the coloured iron(II)-1,10-phenanthroline complex was used for analysis of the iron content of the precipitate. 0.1 g of the dried filter cake was dissolved in 10 ml 0.16 M oxalic acid. After dissolution of the residue, the solution was filled up to 100 ml.

A volume of 5 ml was pipetted into a 100 ml volumetric flask, 5 ml of buffer (40 g NH_4OAc , 50 ml HOAc in 100 ml), 2 ml NH_4OH (100 g L^{-1}) and 2 ml 1,10-Phenanthrolinechloride (5 g L^{-1}) were added and the solution filled to 100 ml. Absorbance was measured at 510 nm in a 10 mm cuvette (Hitachi, U-2000, double-beam spectrophotometer).

2.6. Dyeing experiments

A mass of 9 g washed and bleached wool yarn, metrical number 28/2 m/g, ready to dye was dyed in form of small hanks (Schoeller Wool, Hard, Austria).

Dry dyestuff (0.4–0.5 g) was dissolved during 5–10 min with 15–20 ml 0.08 M oxalic acid at ambient temperature, then 170 ml of warm deionised water were added and the mixture was allowed to rest for 20 min, for completion of dyestuff dissolution.

The wool sample then was added to the dyebath and dyeing was performed using a liquor ratio of 1:20–1:22 at 95 °C in open beakers with manual agitation of the material. A meta-mordating procedure was used. $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ solution 0.18 M (50 g L^{-1}) directly was added to the dyebath, to obtain a final concentration of 0.018 M (5 g L^{-1}) $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$. Sodium acetate 6.1 mmol (0.5 g) was added to increase pH and establish a buffer acetate/acetic acid buffer system. Dyeing was continued for 35 min at 90 °C and the bath was allowed to cool down for 10 min to approximately 60 °C. Then the samples were rinsed 3 times in cold tap water and dried at ambient temperature.

For reference dyeings plant extraction was performed at a liquor ratio of 1:20 and the extract directly was applied using a liquor ratio of 1:20. After 10 min dyeing at the boil, 10% of the bath volume $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ solution 0.18 M (50 g L^{-1}) or $(\text{NH}_4)_2\text{Fe}(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$ solution 0.13 M (50 g L^{-1}) was added and dyeing was continued for 35 min at 95 °C. Dyebaths were allowed to cool down for 10 min, then samples were rinsed in warm tap water and air dried.

The CIELab coordinates and K/S values of the dyeings were measured using a Konica Minolta Spectrophotometer CM-3610d (sample diameter 8 mm, geometry d/8°, Konica, Japan). The L^* , a^* b^* values were calculated for illumination D65.

3. Results and discussion

3.1. Extraction of colourants

Representative dye molecules found in Canadian Goldenrod belong to the group of flavonoids e.g. quercetin (C.I. 75670, Natural Yellow 10, 13), quercitrin (C.I. 75720, Natural Yellow 10), isoquercitrin, rutin (C.I. 75730, Natural Yellow 10) and kaempferol (C.I. 75640, C.I. Natural Yellow 13,10) [1,2,23].

These dyes can be transferred into the aqueous phase by hot water extraction. The release of colouring matter can be monitored

by direct measurement of the absorbance and by determination of the TPH in the extract and by HPLC methods [23]. From literature it is known that rutin, the quercetin-3-rutinoside constitutes the major fraction of extractable yellow chromophores. During the hot extraction and the following dyeing process the major part is assumed to hydrolyse to release quercetin Fig. 1.

The absorbance of an extract at the end of the 60 min extraction process is shown in Fig. 2.

The spectra shown in Fig. 2 demonstrate the reproducibility of the extraction procedures. Only minimal differences were obtained between three independent extraction experiments. The low absorbance in the range of 650–750 nm indicates presence of low amounts of turbidity forming material in the filtered extracts.

To evaluate the required extraction time to reach extraction equilibrium and to check the procedure for eventual decomposition of dyestuff, the absorbance and TPH content of the extract was measured as function of time.

In Fig. 3 increase in absorbance of the aqueous extract measured at 400 nm and the corresponding TPH concentration calculated as gallic acid monohydrate equivalents are shown as function of extraction time.

The absorbance with extraction time reached a maximum after approximately 40 min of heating. Similar results are found for the TPH concentration in the extracts. The results indicate that under the given experimental conditions an optimum concentration already is reached after 30–40 min extraction time. Prolonged extraction does not result in further increase in concentration of coloured matter, also the TPH concentration stabilises. The slight decrease in both, absorbance and TPH concentration indicates that losses of colouring matter occur when extraction time is prolonged substantially over 60 min.

3.2. Precipitation and preparation of solid dyestuff

A considerable part of the flavonoid dyes is able to form complexes with iron(II)-ions [19,20]. Preferred binding sites involved in iron chelation are the 3-hydroxy and 4-oxo groups or the 5-hydroxy and 4-oxo groups [18,19].

The formation of the iron complex is accompanied with a distinct change in colour. While the flavonoid extracts from Canadian Goldenrod appear yellow, the corresponding iron complexes exhibits dark olive colour [15]. The low solubility of the iron-flavonoid complexes can be used to precipitate a concentrated

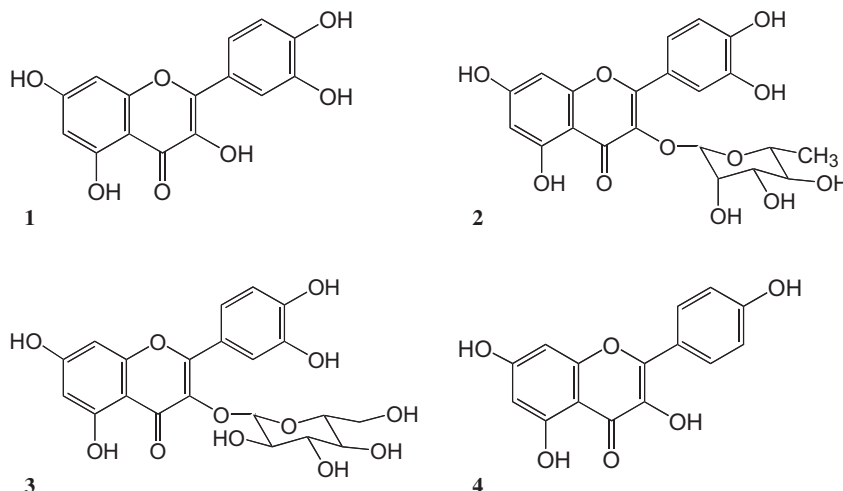


Fig. 1. Representative structures of natural colourants present in Canadian Goldenrod: quercetin 1, quercitrin 2, isoquercitrin 3, kaempferol 4.

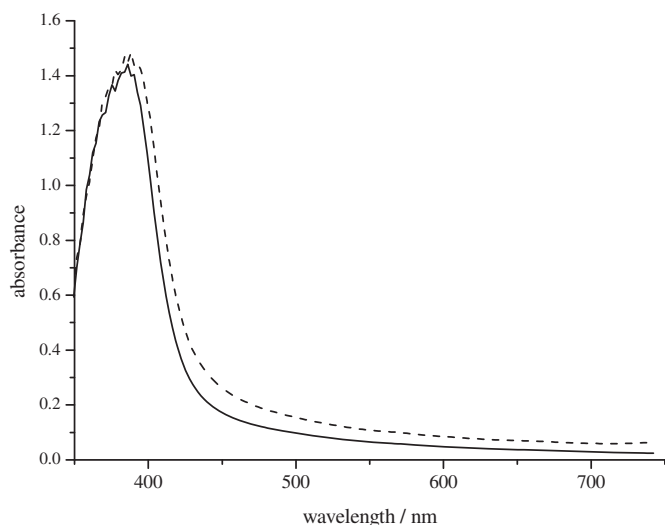


Fig. 2. Absorption spectra measured at two representative filtered aqueous extracts from Canadian Goldenrod (1 g plant material extracted in 10 ml water, 60 min at 60 °C, 1 ml extract diluted with 10 ml water).

dye lake from the aqueous extract and to isolate a solid dyestuff product [14]. The use of iron-salt to precipitate the colourant is consistent with the later use of the material in dyeing processes, where iron mordants are applied. Also acceptable light fastness was reported for dyeings with use of Canadian Goldenrod and iron mordanting [8].

The production of the lake will be performed at the site of farming/harvesting, thus simple procedures were chosen, which can be handled by farmers without high investment.

After pH adjustment of the extract to pH 7.0 precipitation of the flavonoid-iron complex was initiated by addition of 25–200 ml L⁻¹ of a solution containing 0.18 M (50 g L⁻¹) iron(II)sulphate-heptahydrate. Dependent on the added volume of iron(II)sulphate solution the final concentration of iron(II)sulphate in the extract was between 4.39 mM (1.22 g L⁻¹) and 30.0 mM (8.33 g L⁻¹) iron(II)-salt.

The pH in the solution decreased with amount of added iron(II)-salt. At a concentration of 4.39 mM (1.2 g L⁻¹) iron(II)sulphate a pH of 5.7–5.8 was measured in the mixture, while in presence of 30 mM (8.33 g L⁻¹) iron(II)salt a pH of 4.37 was observed.

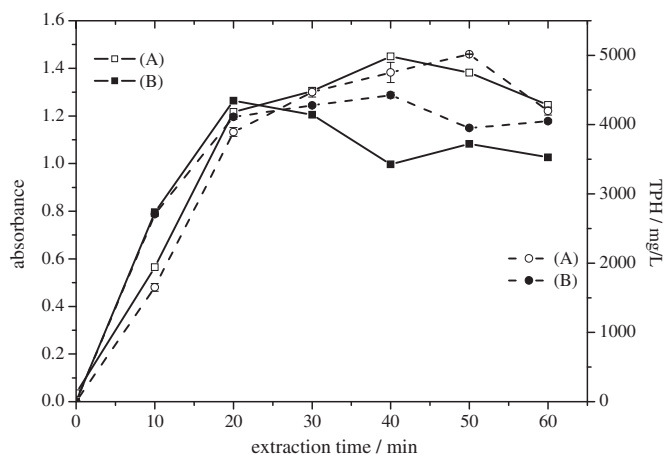


Fig. 3. Absorbance (400 nm) of Canadian Goldenrod extract and TPH content as function of extraction time; extraction A (□) absorbance, (○) TPH; extraction B (■) absorbance, (●) TPH.

The precipitated extract gently was stirred for 2 h to grow particle size and then was allowed to settle overnight. The precipitate was collected by filtration through a paper filter and dried at 60 °C and weighted. In Table 1 the mass of precipitate collected as function of added iron(II)sulphate is shown for representative experiments.

From extraction of 20 g plant material with 200 ml distilled water a mass of 0.6 g–1.1 g dry filter cake was obtained. An increase in concentration of FeSO₄·7H₂O from 4.39 mM (1.22 g L⁻¹) to 8.56 mM (2.38 g L⁻¹) did not increase the amount of dyestuff precipitate to the same extent. At a concentration of 30.0 mM (8.33 g L⁻¹) Fe-salt the mass of dry residue increased only by approx. 70%. As an average value a mass of 30–40 g dry precipitate can be obtained per kg of extracted dry plant material.

To analyse the precipitates in more detail, the iron content and the TPH content of the dyestuff products were analysed. The dried material was dissolved in oxalic acid and after appropriate dilution the iron content was analysed by complex formation with 1,10-phenanthroline and measurement of absorbance at 510 nm. TPH analysis was performed using the Folin-Ciocalteu method.

The unspecific precipitation of iron-hydroxides and other iron containing organic residues increases with concentration of FeSO₄·7H₂O added. As a result the iron content in the dry residue increases from 4.0% after precipitation in presence of 4.39 mM Fe (expt. 1–3) to 6.3% when a concentration of 30 mM Fe was adjusted (expt. 6). Parallel the content of TPH in the dry residue lowers from 50% when a concentration of 4.39 mM iron-salt was used (expt. 1–3) to 40% when precipitation was initiated by 30 mM iron-salt (expt. 6).

The results indicate that a concentration of 4.39 mM (1.22 g L⁻¹) FeSO₄·7H₂O already is sufficient to precipitate the major part of the colourant. Higher FeSO₄·7H₂O concentration did not yield substantially higher amounts of phenolic components in the residue. When higher amounts of precipitate were obtained, this increase was compensated by a lower TPH content in the filter cake.

The use of 4.39 mM (1.22 g L⁻¹) FeSO₄·7H₂O is also favourable with regard to the efficiency in FeSO₄·7H₂O consumption. From the amount of iron used for precipitation and the iron content in the residue, efficiency with regard to iron balance can be calculated. 50% of the added iron had been precipitated at a concentration of 4.39 mM (1.22 g L⁻¹) FeSO₄·7H₂O. Less than 20% of the iron added, were collected in the precipitate, when a rather high amount of 30.0 mM (8.33 g L⁻¹) FeSO₄·7H₂O had been used (Table 1).

In a similar approach the efficiency of TPH precipitation can be estimated. As an average value for TPH concentration in the extract a concentration of 4500 mg L⁻¹ TPH was found (Fig. 2). In 200 ml extract thus a total amount of 900 mg phenolic compounds as gallic acid monohydrate equivalents is present (Fig. 3).

In the precipitation experiments as an average value 0.7 g residue with a TPH content of 45% was obtained. This corresponds to an absolute mass of 315 mg TPH. From the extracted amount of

Table 1

Mass of dry precipitate isolated from 20 g of plant material as function of iron(II)-salt concentration used, pH of solution. Fe-content and TPH of the dry residue; Fe-content in residue in % of total iron added for precipitation.

No.	c(FeSO ₄ ·7H ₂ O)		pH	m of precipitate		Fe-content of plant material	Fe-content	TPH content	Fe in residue
	g L ⁻¹	mM		g	%	%	%	%	%
1	1.22	4.39	5.78	0.64	3.20	4.53	50.4	57.8	
2	1.22	4.39	5.72	0.64	3.20	4.04	47.4	51.5	
3	1.22	4.39	5.53	0.36	1.80	4.41	47.6	31.6	
4	2.38	8.56	5.43	0.69	3.45	5.39	44.5	37.0	
5	2.38	8.56	5.37	0.75	3.75	5.55	45.4	41.5	
6	8.33	30.0	4.37	1.12	5.60	6.32	40.4	17.6	

900 mg TPH a share of 35% could be precipitated as dyestuff containing residue. From 1 L of aqueous extract (TPH 4500 mg L⁻¹) an amount of 1575 mg TPH will precipitate after addition of FeSO₄·7H₂O solution.

In the literature the concentration of phenolic compounds extracted from *Solidago canadensis* in aqueous and alcoholic medium has been analysed by HPLC methods [12]. From the analysis of the phenolic compounds in the aqueous extracts a concentration of 559 mg L⁻¹ rutin, 78 mg L⁻¹ isoquercitrin and 110 mg L⁻¹ quercitrin were reported and a total concentration of phenolics of 747 mg L⁻¹ was found. In this study extraction of the dry plant material was performed 1:10, compared to 1:40 in [23]. Thus an approximately 4 times higher concentration of coloured phenolic compounds in the order of 2900 mg L⁻¹ could be expected in the extract based on reference [23]. In this study approximately a TPH concentration of 4000 mg L⁻¹ TPH was determined, which can be explained with the longer extraction time of 60 min, used in this study.

3.3. Dyeing experiments

To investigate the potential of the precipitated dye for dyeing purposes, exhaust dyeings under standardised conditions were performed using wool as substrate.

For these experiments residues, which had been precipitated with different amounts of FeSO₄·7H₂O were used. The dyeings were evaluated by measurement of the CIEL*a*b* coordinates, the measurement of the diffuse reflectance between 400 and 700 nm and calculation of the Kubelka-Munk value K/S at 400 nm (Table 2). The diffuse reflectance of dyeings 1–4 (Table 2) in the wavelength interval 400–700 nm is shown in Fig. 4

For an assessment of colour depth the respective amount of plant material used has been calculated for the different dyeings in Table 2, an average amount of 0.65 g of dye precipitate per 20 g extracted plant material has been used for the calculation.

Reference dyeings with direct use of plant extracts and CIELab values from literature both for using iron mordanting are given in Table 2 for comparison.

The equivalent amount of dye precipitate used for test dyeings 1–4 corresponded to 15–17 g plant material per 10 g wool, thus darker dyeings were obtained when compared to literature results where the plant extract was used directly (dyeing 7, 8). Reference dyeings obtained with another Canadian Goldenrod plant material showed very similar results in terms of L*, a*, b* values (dyeing 5,6). The differences between reference dyeings 5,6 and 7,8 also demonstrates the high variability in colour strength between

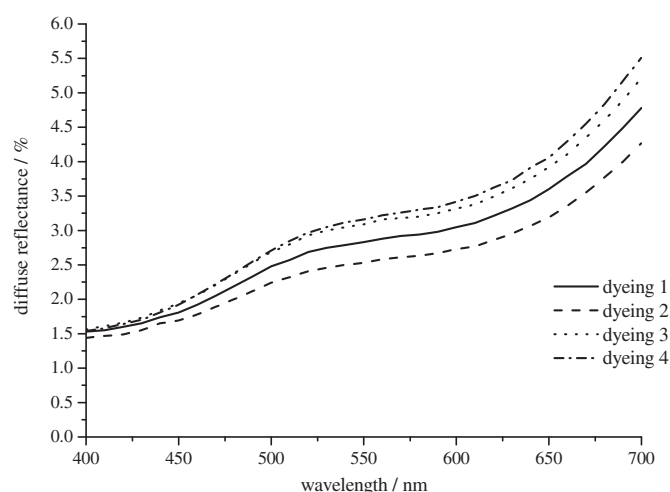


Fig. 4. Diffuse reflectance curves of the test dyeings 1–4 (Table 2).

different plant sources. This variability will be overcome by use of a concentrated product which can be standardised more easily [14].

The diffuse reflectance curves of the samples are shown in Fig. 4.

4. Conclusions

There is significant interest to re-introduce natural dyeing into present technical production scale for textile colouration. For the extraction of plant material a huge amount of plant material has to be handled in the dyehouse, which is a major drawback for a wider future use of such dye sources. The production of a concentrated dyestuff containing product was achieved by use of simple methods, which contain extraction, precipitation, sedimentation, filtration and drying. The processes require only basic equipment and thus can be installed at the site of farming and harvesting. This technique has significant potential to replace the direct extraction of plant material in the dyehouse. In addition quality standardisation for improved reproducibility of the colour depth and shade can be done at the stage of dyestuff production.

The mass of dry residue that can be produced per kg of plant material is limited by the rather low dyestuff content present in the plant. In the given examples an average value of 3–4% of the dry plant mass was found to be formed by addition of FeSO₄·7H₂O solution. A concentration of 7.2 mM (2 g L⁻¹) FeSO₄·7H₂O was found to be sufficient to form a solid precipitate with an Fe-content of 5% w/w and 45% w/w TPH (as gallic acid monohydrate equivalents).

The solid dyestuff is soluble in diluted acid and yields dyeing results which are comparable to the dyeings obtained with direct use of plant material extracts.

Highly concentrated dyestuff solutions up to 100 g L⁻¹ dyestuff are required for continuous dyeing procedures. At present the low concentration of dyestuff in the extract limits the possible application techniques for dyehouses, thus mainly exhaust dyeing procedures are used. This limitation can be overcome when concentrated dyestuff product is available, which permits production of higher concentrated dye solutions and thus allows application in a wide range of different dyeing techniques.

Acknowledgement

Authors thank the FFG (Österreichische Forschungsförderungsgesellschaft) for financial support of the project 814972 Colors of Nature – Pflanzenfarbstoffe in der Praxis, which has been funded in the programme line Factory of the Future.

Table 2

Dyeing experiments obtained with different precipitates, mass of dyestuff and equivalent plant material per 10 g of dyed wool, CIELab coordinates; reference experiments were performed with direct use of extracts.

Dyeing	Precipitation c(FeSO ₄ ·7H ₂ O)		Dyeing m m (dyestuff) (plant material)		L*	a*	b*	K/S (400 nm)
	g L ⁻¹	mM	g	g				
1	4.54	16.33	0.55	16.9	19.21	0.40	7.54	31.00
2	3.49	12.55	0.52	15.0	17.96	0.45	6.66	32.92
3	2.38	8.56	0.48	14.7	20.23	0.29	8.11	30.51
4	1.22	4.39	0.56	17.1	20.50	0.45	8.67	31.00
5 Extract	—	—	—	10	21.90	0.53	7.05	—
6 Extract	—	—	—	10	22.96 ^a	0.49	7.87	—
7 ref. [8]	—	—	—	10	27.05	-0.75	8.43	—
8 ref. [15]	—	—	—	10	28.1	-0.4	11.5	—

Raw wool: L* 83.81, a* -0.05, b* 11.04; K/S 0.49.

^a use of (NH₄)₂Fe(SO₄)₂·6H₂O as mordant.

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