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NATURAL DYE EXTRACTION FROM *PELARGONIUM ROSEUM* PLANTS USING ULTRASOUND METHOD

BY

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Abstract. Dyes derived from natural sources have been used since ancient times in households and in various fields. Subsequently, synthetic dyes were synthesized, but in terms of toxicity, the variant in which the existence of substances harmful to the environment is not a problem is the first category. The aim of this paper is to discover a better solid-liquid extraction technique to avoid the infiltration of dyes found in groundwater following the discharge of wastewater in which they are contained. A source of natural dyes is represented by plant materials that are used in plant research applications, in food, as well as in the paint industries. These could be Green wattle bark, Marigold flowers, Pomegranate rinds, *Pelargonium roseum*, 4’o clock plant flowers, Cocks Comb flowers and more like these. We used *Pelargonium roseum* plants. The influence of ultrasound on the extraction of natural dyes from this material plant as a source of dyes has been studied in comparison with the process of magnetic stirring. Analytical studies, such as UV-VIS spectrophotometry and gravimetric analysis, were performed on the extract. In this regard, the present paper aims at screening the locally available natural dye yielding plants for dyeing purpose. The effect of power ultrasound in the extraction of natural dyes has been studied. Improving the efficiency of dye extraction from **plant materials** with **87.5%** by using ultrasound has led to the conclusion that this method is much faster and more efficient than magnetic stirring.

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Keywords: *Pelargonium roseum*; dye solid–liquid extraction; ultrasound sonochemistry.

1. Introduction

The starting idea of this work is the extraction of dye from the *Pelargonium roseum* plant for dyeing textile substrates and other materials (Sibiescu and Vizitiu, 2015).

There is a need for innovative techniques to improve the major mechanism of natural dye extraction such as rupture the cell wall, release of natural dye and improve the transport of dye in the external medium represented by the solvent.

A solid–liquid leaching process involving mass transfer, represents the extraction of coloring matter.

Use of power ultrasound is classified according to frequency range as: power ultrasound (20–100 kHz) and diagnostic ultrasound (1–10 MHz).

When a liquid is irradiated by ultrasound, microbubbles which appear, grow and oscillate extremely quickly and even collapse violently if the acoustic pressure is high enough. Moreover, in the liquid phase surrounding the particles, high micromixing will increase the heat and mass transfer and even the diffusion of species inside the pores of the solid (Contamine *et al*., 1994).

Ultrasound has been used as a process intensifier for various unit operations in leather processing (Sivakumar *et al*., 2001, 2007, 2009a, 2009b, 2009c) such as leather dyeing, essentially to enhance the diffusion of chemicals through skin/leather matrix.

The use of electric pulse studied in the process (Fincan *et al*., 2004) may involve operational difficulties.

To improve the extraction efficiency the studied gamma ray irradiation technique led to possible degradation and instability of coloring matter (Nayak *et al*., 2006).

2. Experimental Methods

2.1. Experimental Setup

Ultrasonic extraction experiments were performed using Ultrasonic cleaner CD 4820 2500 mL (20 kHz and 0–400 W) in a glass vessel with provisions to set required output power and time as shown in Fig. 1. This device is also equipped with a magnetic stirrer and a temperature controller.

Fig. 1 – Ultrasonic cleaner CD 4820.

2.2. Materials and Methods

2.2.1. *Natural Dye Materials*

We used *Pelargonium roseum* flowers, as plant material containing natural dye. These plant materials were collected fresh from Iaşi - Botanical Garden and are shown in Fig. 2.

Fig. 2 – *Pelargonium roseum* flowers.

2.2.2. *Extraction Using Magnetic Stirring (Control Experiment)*

For the experiment, fresh pink *Pelargonium roseum* flowers petals were collected from the Botanical Garden. 1 g of 1 cm sample was used and 50 mL of distilled water was added to a Berzelius glass to keep the plant materials together with the tip of the ultrasound completely immersed in the solvent.

The sample with petals and water was stirred magnetically for 3 hours. To prevent solvent loss by evaporation the Berzelius glass was coated with aluminum foil.

The temperature of the extraction bath for the control process was also maintained at 45°C.

The optical density of the extract samples, taken every 30 minutes, was determined using the S22 UV / VIS Spectrophotometer. The yield and efficiency of extraction of each sample were determined by the gravimetric method at the end of 3 hours. The extract was sealed and stored at low temperature for future reference.

2.3. Analytical Methods

2.3.1. *Spectrophotometric Analysis*

The extract samples of *Pelargonium roseum* were analyzed.

The intense pink color dye present in the extracted solution was analyzed using a spectrophotometer UV-2101PC in the visible region of 400–800 nm, after suitably dilution. This was made by measuring the absorbance value at a maximum specific wavelength.

2.3.2. *Gravimetric Analysis*

The equation used for calculating the yield was:

$$\% \text{ yield of natural colorant} = \frac{\text{natural dye extract obtained (g)}}{\% \text{ amount of plant material used (g)}} \quad (1)$$

$$\% \text{ improvement due to ultrasound} = \frac{\% \text{ yield of (ultrasound process-control)}}{\% \text{ yield of control}} \quad (2)$$

At the end of each extraction process, the samples taken from both ultrasound and control extracts were filtered. The extracts were dried in a hot-air oven until all the water was evaporated and only the extract remained. The samples from the weighting bottle were then cooled in a desiccator and weighted. This procedure was repeated until constant weight.

The weight of the colorant extract obtained per gram of the plant material used, was calculated to determine the percent yield of natural colorant using the above Eq. (1).

3. Results and Discussions

UV–VIS spectrum of natural dye obtained from *Pelargonium roseum* was obtained. The dye was extracted from the petals of this plant and analyzed at the wavelength of 670 nm. The extraction of the natural dye from the

Pelargonium roseum was performed from intense pink petals using water as solvent.

The absorbance values for natural dye extract obtained by ultrasound and magnetic stirring control are shown in Fig. 3 (*a*, *b*).

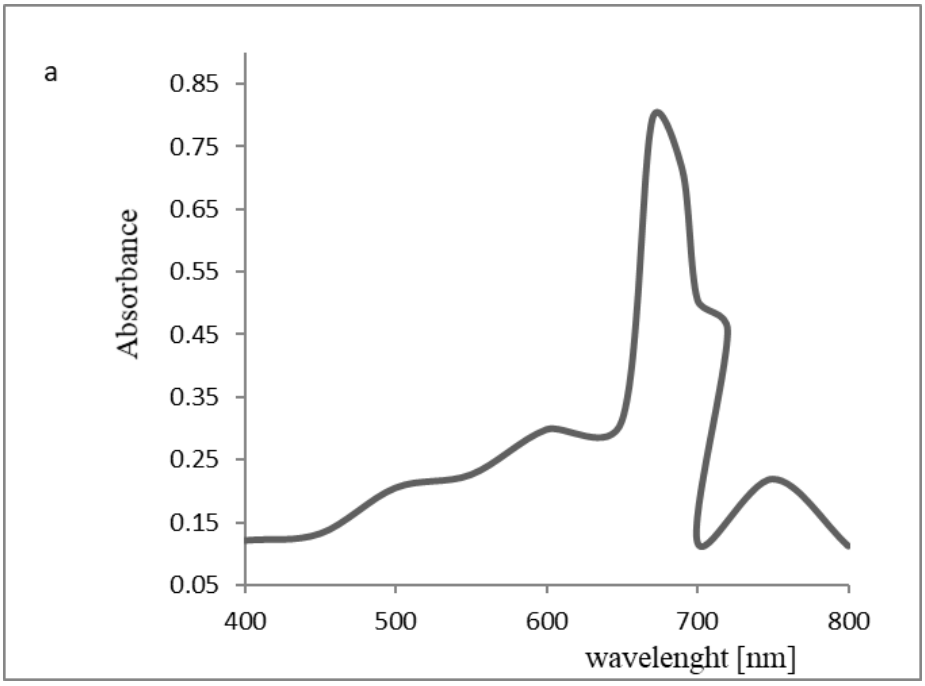
The results indicate that there is about 87.5% improvement in the % yield of extract due to the use of ultrasound as compared to the control process as shown in Table 1. The difference in increasing the ultrasonic extraction yield could be due to the different degree of binding of the dye attached to the cell membranes of the plant.

Another important factor are the chemical components present in the pink petals of the *Pelargonium roseum* responsible for the color (chromophore group) and the nature of their solubility. These aspects are planned for our future study.

Table 1

The Effect of Ultrasound on the Obtained Yield

Magnetic stirring control – yield (%) (A)	Ultrasound – yield (%) (B)	%Improvement due to ultrasound, ((B - A)/A) × 100
16	30	87.5



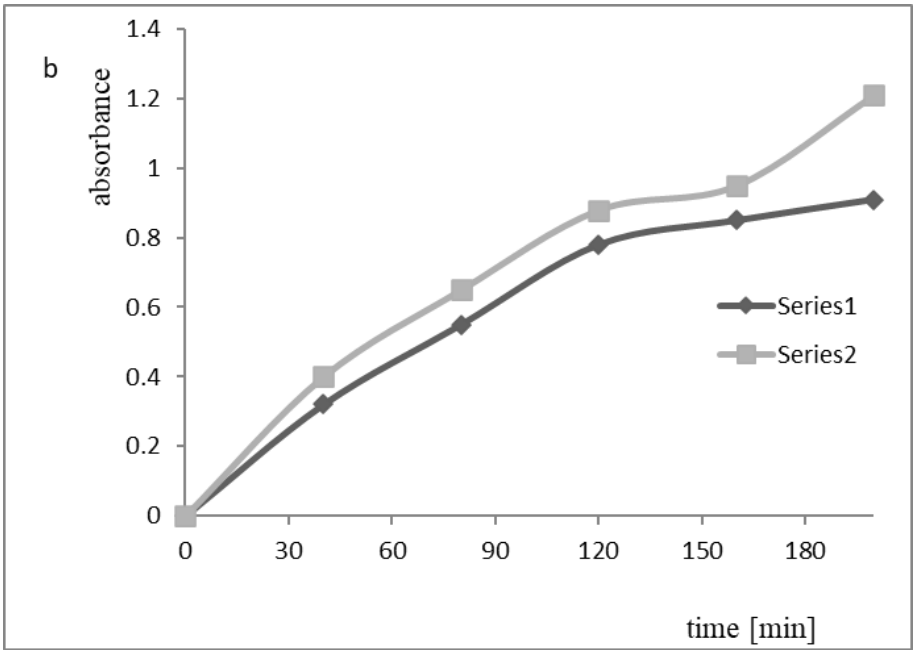


Fig. 3 – (*a*) UV-VIS spectrum for *pelargonium roseum*; (*b*) The absorbance values at maximum wavelength 670 nm for natural dye extract obtained with magnetic stirring (series 1) and by ultrasound (series 2).

4. Conclusions

Natural dyes provide a safe environmentally friendly option for coloring food and other materials. It has been found that the application of ultrasound can increase the extraction of dyes from different parts of various plant resources.

The extraction was done using ultrasound as well as magnetic stirring methods and the kinetics and efficiency of the extraction were compared. The reason for the improvement could be due to a better leaching of the natural dye material from the plant cell membranes and the mass transfer in solvent assisted by the acoustic cavity provided by ultrasound.

The results indicate that there is an improvement of 87.5% yield of the extract obtained due to the use of ultrasound compared to magnetic stirring at 45°C. Various process parameters, such as solvent system, temperature, ultrasonic power, amount of coloring material etc. are of interest for our study as future works.

We would expect better extraction efficiency with solvents such as n-hexane for those dye materials more soluble in organic solvents. But our goal is

to develop an efficient sustainable process with an aqueous system without using organic solvents.

Extraction efficiency may decrease if the temperature is below 45°C; however, higher temperatures could affect the plant material itself as they are sensitive to the same.

This new technique can be used effectively for the extraction of dyes from various plant resources, even without the conventional heating required conditions. This process ensures the efficient use of natural resources as an ecological method in the current situation of global environmental concern.

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EXTRACȚIA COLORANTULUI NATURAL DIN MATERIALUL VEGETAL *PELARGONIUM ROSEUM* FOLOSIND METODA ULTRASONĂRII

(Rezumat)

Coloranții proveniți din surse naturale au fost utilizați din cele mai vechi timpuri în gospodării şi în diverse domenii.

Ulterior, au fost sintetizați coloranți sintetici, dar din punct de vedere al toxicității, varianta în care nu există substanțe nocive pentru mediu înconjurător, firește o reprezintă prima categorie.

Scopul acestei lucrări este de a descoperi o tehnică mai bună de extracție solid-lichid, pentru a evita infiltrarea coloranților care se găsesc în apele subterane, în urma evacuării apelor uzate în care sunt conținuți. O sursă de coloranți naturali o reprezintă materialele vegetale care sunt utilizate în mai multe domenii: medicină, în produse alimentare, precum și în industria textilă, în vopsitorie.

Acestea ar putea fi: scoarță de acacia, gălbenele, sunătoare, coajă de rodie, *Pelargonium roseum* (muşcata roz), Creasta cocoşului şi altele asemenea.

În această lucrare s-a lucrat cu petalele plantei *Pelargonium roseum*.

Influența ultrasunetelor asupra extragerii coloranților naturali din petalele acestei flori (posibilă sursă de coloranți) a fost studiată în comparație cu procesul de agitare magnetic, calculându-se în fiecare caz, randamentul.

Din datele experimentale s-a concluzionat că procedeul de extracție a colorantului prin ultrasonare este mult mai bun decât cel prin agitare magnetică.