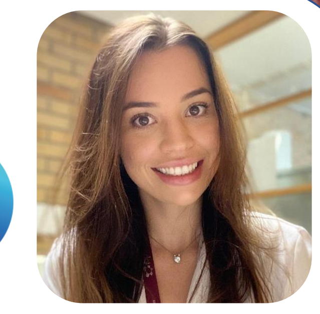

Eco-Friendly Coloration of Leather with Natural Dyes: Performance Insights and Industrial Validation

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INTRODUCTION

The transition to more sustainable materials is a growing priority in the leather industry, particularly regarding the environmental impact of synthetic dyes. Natural dyes, derived from renewable sources, present an eco-friendly alternative that can reduce environmental pollution and support cleaner production practices [1]. They are biodegradable, non-toxic, and non-carcinogenic; however, their widespread adoption is still limited by challenges including poor colour fastness, low substrate affinity, and variability in shade reproducibility [1,2]. The ongoing research aims to address these limitations, enabling the leather sector to combine sustainability with high performance through greener ingredients and technologies [2].

OBJECTIVE

This study aims to evaluate and compare the dyeing performance of several natural colorants applied to wet-white leather, from laboratory trials to preliminary assessments at semi-industrial scale.

EXPERIMENTAL

Seven natural dyes were selected for evaluation: **BioIndigo** from True Indigo (*Indigofera tinctoria*), **Kareel** from Myrobalan (*Terminalia chebula*), **Leaf Green** from Mulberry (*Morus alba*), **Mallow** from Pomegranate (*Punica granatum*), **Rubia** from *Rubia cordifolia* root, **Nimbus** from the secretion of the *Kerria lacca* insect, and **Yeliona**, a blend of three extracts—Marigold (*Tagetes erecta*), Tesu (*Butea monosperma*), and Kamala (*Mallotus philippensis*).

The mordanting process, also natural, was performed on wet-white leather using 6% OLW (on leather weight) of chitosan (85% degree of deacetylation), prepared in a 0.1 M acetic acid solution. The use of chitosan as a mordant represents a new strategy in leather dyeing, as it has previously been applied mainly to textiles.

The laboratory procedure is illustrated in **Figure 1**. After dyeing, the leather samples were characterized. Colorimetry (CIELAB) was performed to measure L*(lightness), a* (red - green), b* (yellow - blue), and ΔE values, calculated as $\Delta E = \sqrt{(L_2^* - L_1^*)^2 + (a_2^* - a_1^*)^2 + (b_2^* - b_1^*)^2}$. Colour stability was evaluated over time, while colour fastness was assessed for resistance to artificial light, perspiration, and rubbing (in dry, wet, and alkaline conditions).

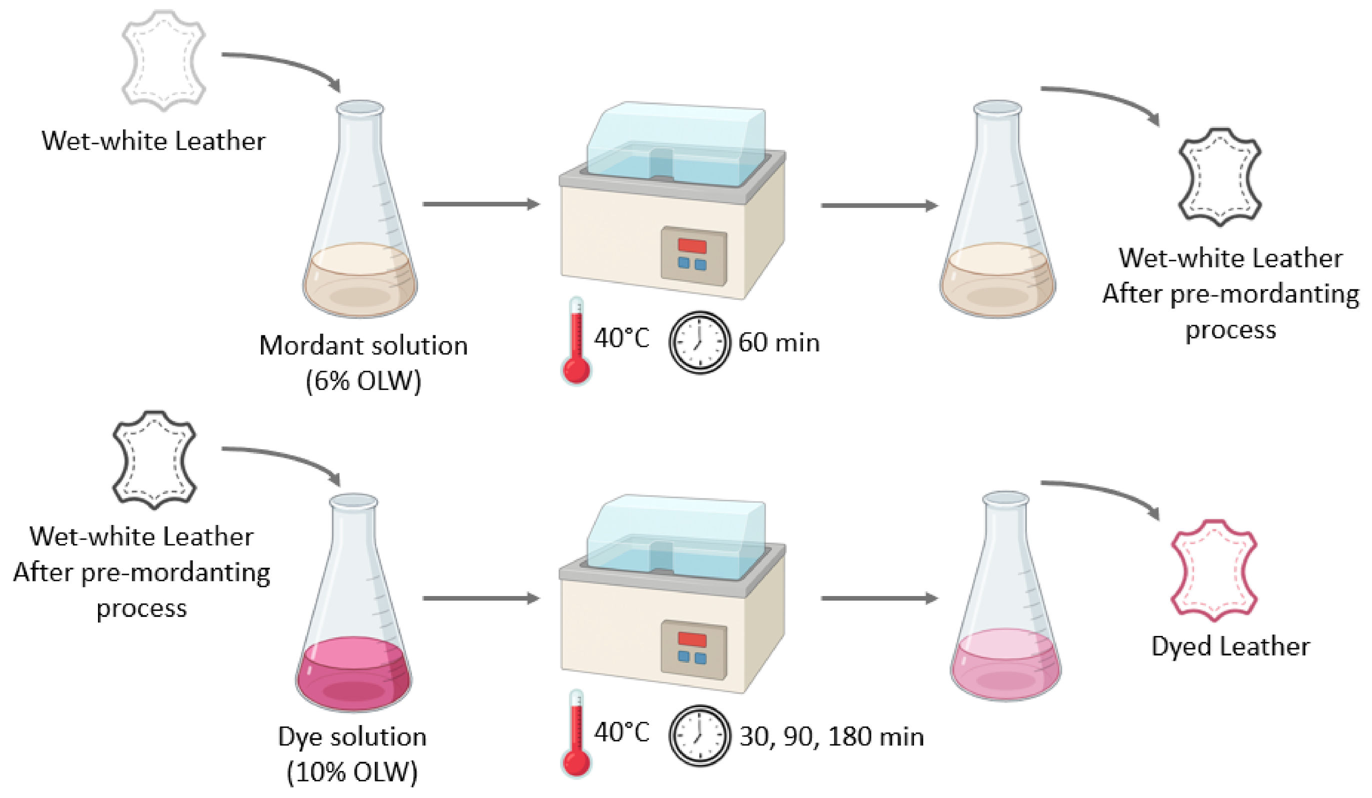

Figure 1. Schematic representation of the methodology for dyeing leather on a laboratory scale.

RESULTS

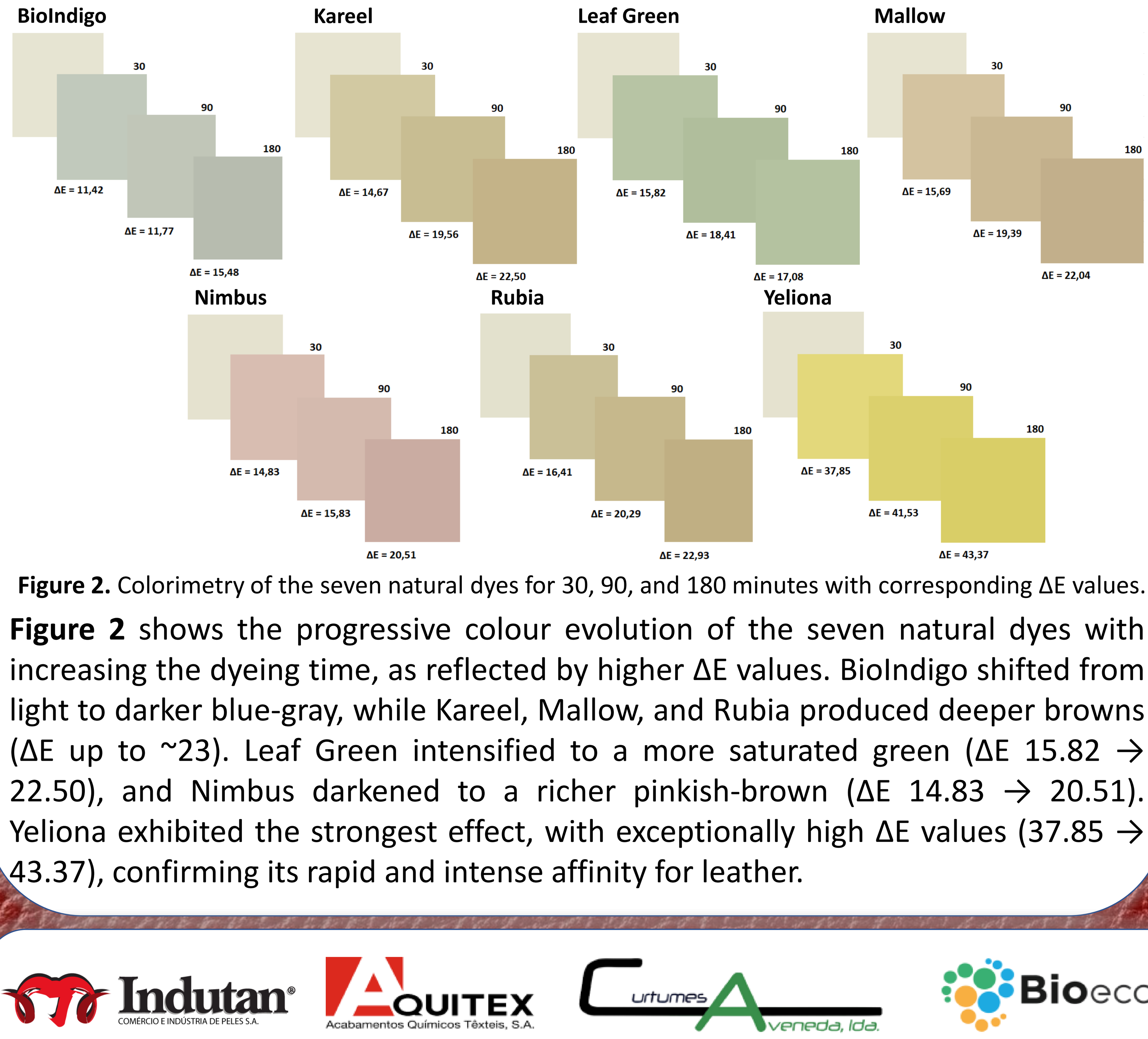

Figure 2. Colorimetry of the seven natural dyes for 30, 90, and 180 minutes with corresponding ΔE values.

Figure 2 shows the progressive colour evolution of the seven natural dyes with increasing the dyeing time, as reflected by higher ΔE values. BioIndigo shifted from light to darker blue-gray, while Kareel, Mallow, and Rubia produced deeper browns (ΔE up to ~23). Leaf Green intensified to a more saturated green (ΔE 15.82 → 22.50), and Nimbus darkened to a richer pinkish-brown (ΔE 14.83 → 20.51). Yeliona exhibited the strongest effect, with exceptionally high ΔE values (37.85 → 43.37), confirming its rapid and intense affinity for leather.

The colour stability of the leather was monitored post-dyeing over 60 days, and results showed no significant variations in ΔE values, indicating promising colour fastness of the dyes.

Table 1. Colour fastness to artificial light, perspiration and dry rubbing in dry, wet and alkaline sweat.

Method	BioIndigo	Kareel	Leaf Green	Mallow	Nimbus	Rubia	Yeliona
Fastness to artificial light (xenon lamp)	4	3	1-2	2-3	2-3	3-4	1
Fastness to perspiration – colour change	1-2	4	2	3	1-2	2	2-3
Fastness to perspiration – staining:							
Dicel	4	3-4	3-4	3-4	2-3	1	2
Cotton	4	2-3	3	3	1-2	2	1
Nylon 6.6	4	2-3	3	3-4	2-3	1	2
Polyester	3-4	3-4	3	4	2-3	2-3	2
Acrylic	3	3-4	3-4	4	2-3	3	2
Wool	4	3-4	3-4	3-4	1-2	1-2	2
Rubbing resistance "Veslic" - degree of staining:							
Fastness to dry rubbing, 50 cycles	1	2-3	3	3-4	4	2	4
Fastness to wet rubbing, 20 cycles	1-2	3-4	3-4	4	4-5	2	4-5
Fastness to alkaline sweat rubbing, 20 cycles	2	3-4	3-4	4	4	3	4
Rubbing resistance "Veslic" - degree of colour change:							
Fastness to dry rubbing, 50 cycles	2	4-5	4	4-5	4-5	3-4	4-5
Fastness to wet rubbing, 20 cycles	2	4	3-4	4	4	2	4
Fastness to alkaline sweat rubbing, 20 cycles	2-3	3	3	4	2	1-2	4

*Values displayed in green indicate that the colour fastness meets or exceeds the leather specifications, whereas values in red are below the required standard.

The colour fastness tests presented in **Table 1** showed that BioIndigo and Rubia dyes did not meet the rubbing resistance specifications. In contrast, the performance of the other dyes (Kareel, Leaf Green, Mallow, Nimbus, and Yeliona) aligned with the established standards, in exception of Yeliona, which failed in the fastness to perspiration (staining) tests. These results can stem from the intrinsic nature of the dyes.

The most promising dyeing formulations developed at the laboratory scale with significant market appeal were chosen (Nimbus, Yeliona, and Leaf Green) and subsequently applied to leather at semi-industrial drum trials (collaboration with an industry partner). To specifically evaluate the performance of the natural dyes, the standard industrial process was kept constant, replacing the synthetic dye for the natural ones, as shown in **Figure 3**.

Figure 3. Semi-industrial scale tests of the natural dyes Nimbus, Yeliona, and Leaf Green, from left to right.

The trials confirmed the positive scalability of the process, particularly regarding dye performance, handling, and sustainability attributes.

CONCLUSIONS

The above findings demonstrated that the selected natural dyes not only meet aesthetic and functional requirements but also align with circular economy principles, offering a viable and eco-friendly alternative to conventional ones. This work contributes to bridging the gap between academic research and industrial application in the pursuit of greener leather production processes.

REFERENCES

[1] S. Thiyagarajan, K. Balakrishnan, S. Tamilarasi, J. Appl. Chem. 8 (2015) 85–89.

[2] P. Brudzyńska, A. Sionkowska, M. Grisel, Materials 15 (2022).

ACKNOWLEDGEMENTS

BBioShoes4All (2375-02/C12-i01.01/2022) funded by the EU/Next Generation EU, in the scope of the Recovery and Resilience Program (RRP), through the "Fundo Ambiental". FCT/MCTES (PIDDAC): CIMO, UIDB/00690/2020 (DOI: 10.54499/UIDB/00690/2020) and UIDP/00690/2020 (DOI: 10.54499/UIDP/00690/2020); and SusTEC, LA/P/0007/2020 (DOI:10.54499/LA/P/0007/2020).National funding by FCT, P.I., through the institutional scientific employment program contract of A. Santamaria-Echart.