Studies Regarding the Use of Senescent Viticultural Material as Colorants in the Plastics Industry

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When we talk about plastic industry, then one of the most important aspects of this trade is giving the plastic its bright and attractive color. As more and more people crave for the "return to nature" touch, pigments extracted from vegetal materials considered as waste have been analysed for dyeing textiles and manufacturing colorants for painting. Senescent leaves of Vitis vinifera L. have been tested for their capability of extracting organic pigments (chlorophyll, carotenoids, xanthophylls and anthocyans). The results show that the potential exists, further research is needed in order to best develop the methodologies needed.

*Keywords: vine-leaves, pigments, vegetal waste, CIE L*a*b* space, grape varieties*

Additives, including colorants, for bioplastics are being widely investigated. Of particular interest are colorants that have no adverse effect on health or the environment and do not compromise bioplastics' compliance with compostability standards. In addition to traditional pigments that can be used in biopolymers, bio-derived colorants [1] based on plants [2] and other renewable materials are now available from multiple companies. Colour compounds made from 100% sustainable natural sources are very earthy and organic looking. While some boast excellent clarity, light fastness may be still the constraint. Current bio-derived colorants include: orange curcuma (root of turmeric spice plant), yellow urucum (tropical flower), green (chlorophyll & other plant sources), carmine red (cochineal insect). The use of anthocyans as natural/food colour additives E 163 [3] was analysed as they proved to be non-toxic and non-pollutant [4]. The purpose of this article is the use of viticultural biological materials, seen as waste, is investigated in order to find possible uses as pigments for different industries. The CIE Lab analysis as well as the total polyohenolic content and anthocyans's content of the vegetal material underline the important quantities of pigments that can be extracted and further introduced in industrial cycles [5].

Experimental part

Materials and methods

The present study analyses the possible uses of viticultural material leaves [2] as biological source for organic pigments and dyes 40 grape varieties (Romanian and cosmopolitan, table grapes and grapes for wine-making) from the Ampelographical Collection of the Faculty of Horticulture of USAMV Iasi were taken into analysis. Harvesting was done at the end of September

Analysing the values of the chlorophyll content index, chromatic parameters, anthocyans content, content of total phenolic compounds, some observations can be made regarding its senescent level, its colorimetric characteristics as well as its pigments variation.

Results and discussions

The chlorophyll content index was determined using a CCM-200 plus - Chlorophyll content meter, instrument that, în a quick and precise manner, evaluates tthe chlorophyll content of whole leaves, compared to the intrusive manner used by Biber (figs. 1 and 2).

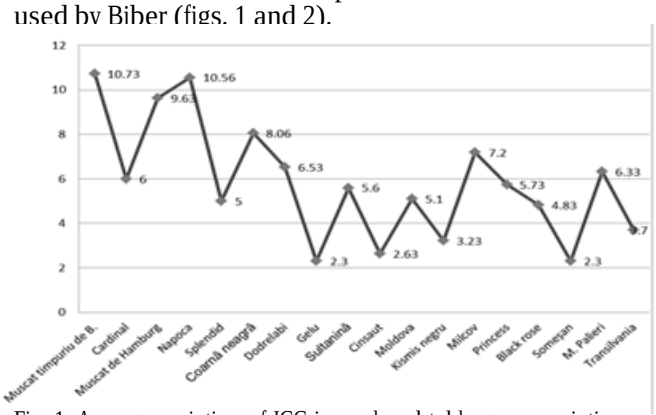

Fig. 1. Average variation of ICC in analysed table grape varieties

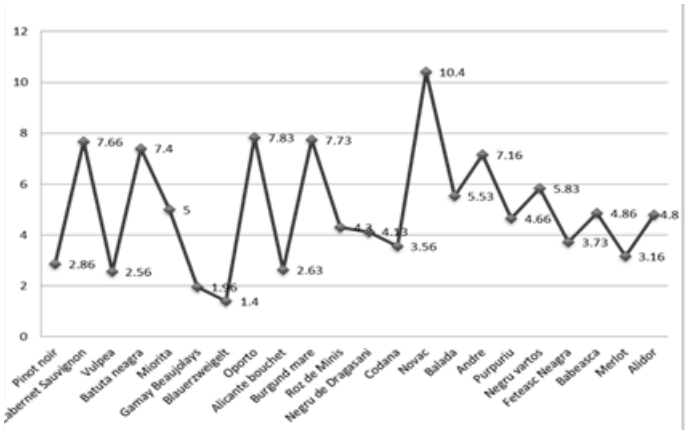

Fig. 2. Average variation of ICC in analysed wine grape varieties

The ICC values of the analysed biologic material varied according to grape variety. The chlorophyll [4] content of the leaves has a peculiar dynamic, with low values during blossoming of the vines, higher values up to the *véraison* moment and again, a big drop in the months of September and October. Previous research [4,6,7] showed that young leaves have a low photosynthetic activity, the pigments concentration reaching a maximum in 20-30 days old leaves.

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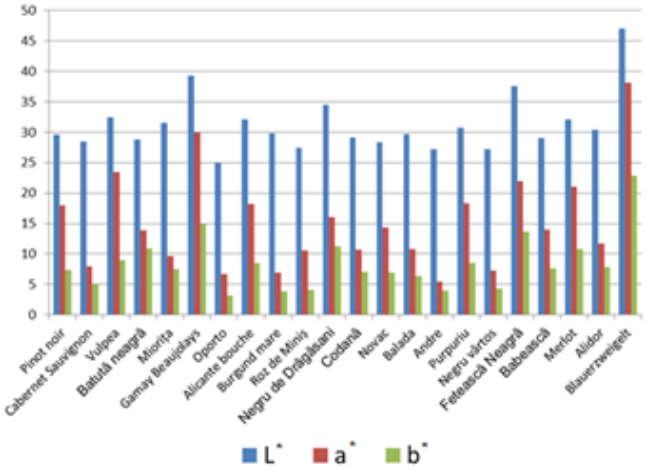

Fig. 3. Variation of chromatic parameters in leaves of analysed wine grape varieties

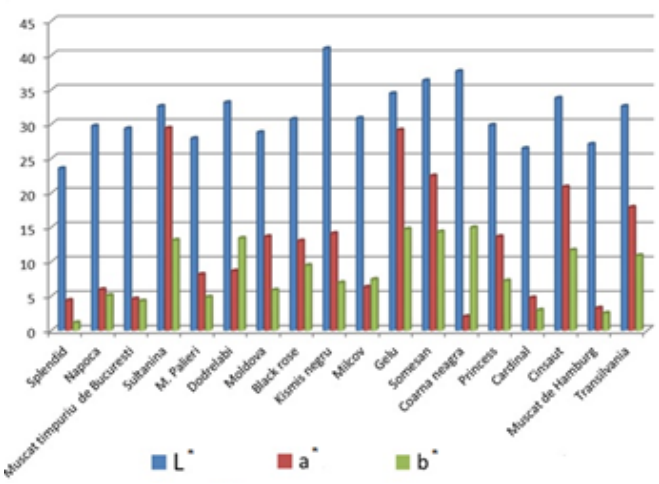

Fig. 4. Variation of chromatic parameters in leaves of analysed table grape varieties

The colour characteristics were determined using a HunterLab MiniScan XE Plus colorimeter. Luminosity L*, parameter a* (red-green) and parameter b* (yellow-blue) were obtained by using a D65 illuminant at an angle of the standard solid of 10°(figs. 3 and 4).

In deep colored extracts, due to a higher phenolic content, luminosity (L*) presents lower values [7].

It can be observed that in all senescent leaves, the chromatic parameter, a* - red-green predominates in all cases, except for the varieties Coarnã neagrã, Dodrelabi and Milcov, where chromatic parameter, *b* – yellow-blue* is highest.

The presence of a bigger quantity of carotenoids and xanthophylls, as well as anthocyans is suggested.

The total polyphenolic compounds as well as anthocyans (table 1) were determined according to OIV-MA-AS2-10, from the International Methods of Analysis Compendium of the O.I.V. [8] while the monomeric anthocyans were analysed according to *p*H variation [9].

Table 1

MONOMERIC ANTHOCYANS CONTENT AND TOTAL PHENOLIC COMPOUNDS OF SENESCENT VINE LEAVES V. VINIFERA L.

Grape variety	(mg EC/100 g)	Total polyphenolic compounds (g EAG/100g)
Pinot noir	245.21	2.79
Splendid	152.35	2.01
Napoca	129.58	1.79
Muscat timpuriu de Bucureşti	148.74	1.98
Cabernet Sauvignon	197.39	2.64
Vulpe	369.81	3.27
Batută neagră	254.01	2.93
Miorița	204.43	2.61
Gamay Beaujolays	410.11	3.44
Sultanină	397.64	3.22
M. Palieri	198.36	2.64
Oporto	178.24	2.47
Alicante bouche	261.90	2.88
Dodrelabi	195.21	2.68
Moldova	224.31	2.71
Burgund mare	179.18	2.39
Black rose	230.67	2.70
Roz de Miniș	209.17	2.51
Negru de Drăgașani	241.06	2.63
Codana	201.34	2.04
Novac	231.71	2.30
Balada	198.19	2.54
Kișmiș negru	230.04	2.81
Milcov	178.09	1.94
Andre	130.44	2.07
Purpuriu	271.65	2.38
Negru vârtos	181.80	1.97
Gelu	389.77	3.14
Someșan	338.13	2.91

Fetească neagră	318.42	2.79
Babească neagră	216.31	2.55
Merlot	297.17	2.73
Coarnă neagră	119.32	1.81
Princess	218.16	2.47
Alidor	187.77	2.02
Cardinal	135.18	2.17
Cinsaut	307.26	2.73
Muscat de Hamburg	111.37	1.77
Blauerzweigelt	417.09	3.31
Transilvania	214.66	2.49

Tabel 1 continuated

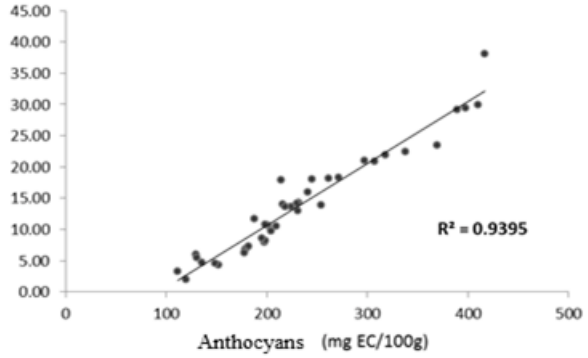

Fig. 5. Correlation between the anthocyans content and the values of chromatic parameter a*

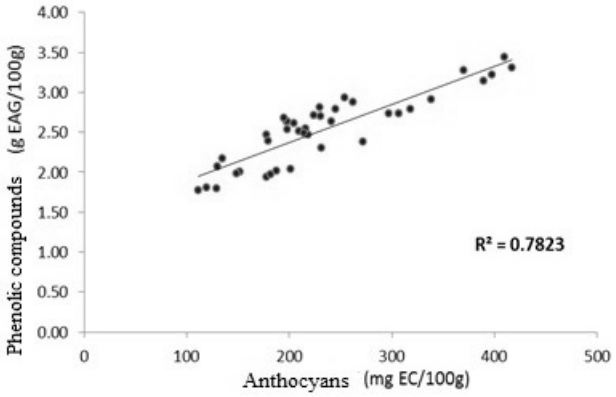

Fig. 6. Correlation of anthocyans content and total phenolic compounds content in senescent *V. vinifera L.* leaves

The possibility of establishing a correlation between the values of chromatic characteristics and the anthocyans' content or total phenols content was analyzed.

An inverse relationship between luminosity and the anthocyans' and phenolic compounds' content was established with low values of the correlation coefficients. A high determination coefficient (R^2= 0.9395) was obtained between the anthocyans' content and the values of the chromatic parameter a*, indicating the positive relationship between the two parameters (fig. 5).

A high content of anthocyans corresponded to a high quantity of phenolic compounds for value R^2 = 0.7823 (fig. 6).

Conclusions

The present study presents the possibilities of using vegetal waste from the vineyard (dry vine leaves, as well as adult green leaves) for their pigment content, in the plastic industry domain. The high number of grape varieties used, both table grapes as well as grapes for winemaking, create a complex background, for researchers to choose from.

The values of the ICC varies according to grape variety, from a minimum of 1.96 in Gamay Beaujolays and a maximum of 10.73 in the Muscat timpuriu de Bucure'ti grape variety.

The anthocyans content in senescent leaves has a maximum in the case of the grape variety Blauerzweigelt (417.09 mg EC/100 g) and a minimum in the case of the variety Muscat de Hamburg (111.37 mg EC/100 g). The same direct relation is available for the total phenolic quantity.

The potential for extracting color pigments from green and senescent leaves exists; however, further research is needed in order to identify the best methodologies for this matter. At the current time, an alcoholic extraction seems to be the best choice, as solvents are not environmentally safe; however, the economic viability of these technologies is to be studied further.

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References

1.PEZZELLA, A., CAPELLI, L., COSTANTINI, A., LUCIANI, G., TESCIONE, F., SILVESTRI, B., VITIELLO, G., BRANDA, F., Materials Science and Engineering Materials for Biological Applications, **33,** 2013, p. 347.
2.VOLF, I., POPA, V., Rev. Chim. (Bucharest), **55,** no. 9, 2004, p. 707.
3.*** Codex Alimentarius, World Health Organization, Food and Agriculture Organization of the United Nation, http://www.codexalimentarius.net/gsfaonline/docs/CXS_192e.pdf accesed on 07/10/2015.
4.KAMAR, F., NECHFOR, A., RIDHA, M., ALTAIEEMI, M., NECHIFOR , G., Rev. Chim. (Bucharest), **66**, no 7, 2015, p. 921.
5.ATODIRESEI, G.V., SANDU, I.G., TULBURE, E.A., VASILACHE, V., BUTNARU, R., Rev. Chim. (Bucharest), **64**, no. 2, 2013, p. 165
6. BURZO, I., Modificări climatice ºi efectele asupra plantelor horticole. Ed. Sitech, Craiova, 2014.
7. LOVISOLO C., SCHUBERT A., RESTAGNO M., Photosynthesis of Grapevine Leaves of Different Age at High and Low Light Intensity, Acta Horticulturae, no.427, 1996, p.171.
8.KELLER M., The Science of Grapevines: Anatomy and Physiology. Academic Press, Elsevier Inc, 2010.
9.*** International Organisation of Vine and Wine , www.oiv.int accessed on 09/10/2015.
10. FILIMON, V., FILIMON, R., ROTARU L., Buletin USAMV Cluj-Napoca, Seria Horticultura, **71** no. 2, 2014, p. 246.
11. TARDEA c. Chimia ºi analiza vinului. Ed. Ion Ionescu de la Brad, Iaºi, 2007.
12.POMOHACI, N., STOIAN, V., GHEORGHITA, M., SIRGHI, C., COTEA, V.V., NAMOLOSANU I., Oenology, Ceres, Bucharest, Romania, 2000.
13.ROTARU, L., Soiuri de vita de vie pentru struguri de vin. Ed. Ion Ionescu de la Brad, Iaºi, 2009.
14. COTEA, D.V., ZANOAGA , C., COTEA, V.V. Tratat de oenochimie, Ed. Academiei Române, Bucuresti, România 2009.

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