Encapsulation of anthraquinone dyes from *Rubia tintorum* L. by freeze and spray-drying for application in the textile industry

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Introduction

Natural dyes have been used since the 1990s to replace synthetic ones due to their biodegradable nature, being anti-allergenic, non-carcinogenic and low toxicity. Nowadays, the dyeing with natural colorant from plants, such as the rubia (*Rubia tinctorum* L.) is carried out at three technological levels, artisanal, semi-industrial and industrial, by companies with good environmental practices. For this, it is essential the stabilization and storage of natural dyes, since they have less stability, to pH, light and washing, when compared with the synthetic ones.

The encapsulation of anthraquinones by freeze-drying and spray-drying was studied on the colour stability, using maltodextrin and Arabic-gum as carrier agents[1]. In the extracts, encapsulated and non-encapsulated, the colour parameters (CIE Lab), total phenolic compounds content (TPC), alizarin content and chemical profile were evaluated by HPLC-DAD combined with HRMS.

Fig. 1 Rubia (*Rubia tinctorum* L.) plant and part of plant used.

Mat & Methods

Anthraquinones compounds were isolated by solvent extraction from dried and powered plants. Freeze-drying and spray-drying were used to produce the water-soluble shell-coated matrix capsules with maltodextrin or Arabic gum as carrier agents. Microcapsules characterization were realized by physical methods and colorimetry assays. The extracts of non-encapsulated and encapsulated anthraquinones compounds were elucidated by Liquid Chromatography and Tandem Mass Spectrometry. Stability tests of the release compounds were done at different pH and at 80ºC. Overall, the results will show the dye plants with higher sources of flavonoid dye compounds, revealing the potential multipurpose use of the rubia plants.

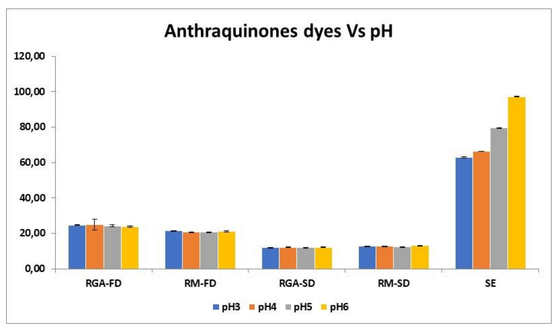
Results

Solubility, encapsulation yeld (EY) and encapsulation efficiency (EE)

The two encapsulation processes, for both encapsulating agents, increase the dispersion of the hydroalcoholic extracts of anthraquinones compounds.
Arabic gum showed lower encapsulation efficiency for both drying processes.

Stability of encapsulated and non-encapsulated anthraquinones in release tests with pH and temperature

The stability of the aglycone anthraquinones to pH variation was greater for the lyophilization process, with no significant differences between the encapsulating agents. Regarding temperature, the stability of anthraquinones is greater when the dye is encapsulated in maltodextrin and no significant differences were observed between the two drying processes.



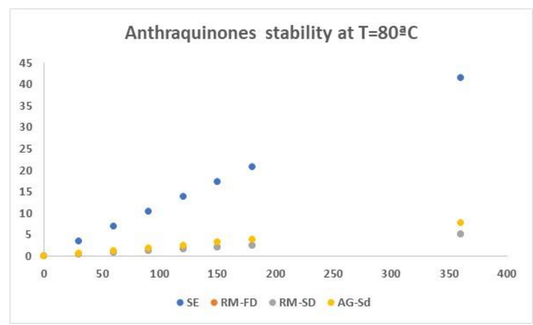


Fig. 2 Stability of non-encapsulated and encapsulated anthraquinones in rubia extract as pH and temperature (T=80°C) function.

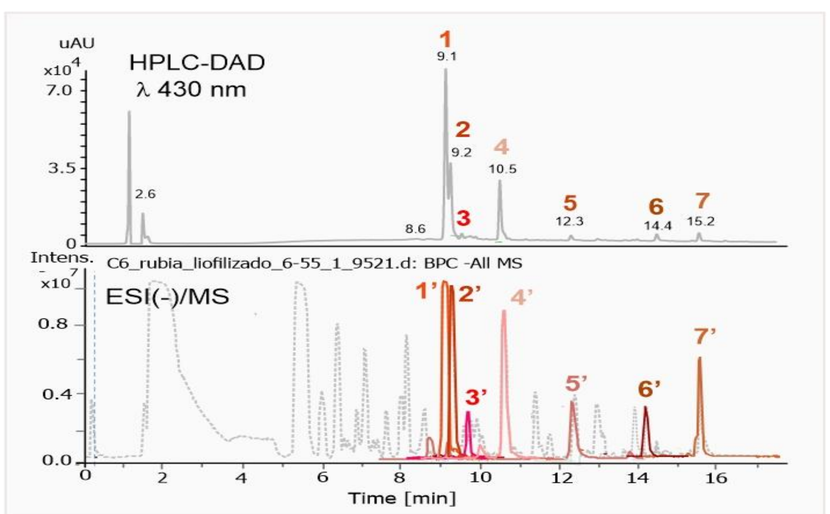


Fig. 3 Chromatographic profile of anthroquinones from rubia extract obtained by HPLC-DAD-HRMS

Peak 1 λ_{max} 266; 406 nm; *m/z* 563.1425 Lucidine primevorisoid
Peak 2 λ_{max} 260; 412 nm; *m/z* 533.1317 Alizarin primevorisoid
Peak 3 λ_{max} 266; 404 nm; *m/z* 431.0994 Lucidine-Glc
Peak 4 λ_{max} 270; 406 nm; *m/z* 547.1473 Rubiadin primevorisoide
Peak 5 λ_{max} 280; 414 nm; *m/z* 269.0464 Lucidine
Peak 6 λ_{max} - 419 nm; *m/z* 239.0359 Alirazine
Peak 7 λ_{max} 278; 410 nm; *m/z* 253.0519 Rubiadine

Red dye profile of rubia (R) extracts

The results allowed the identification of the same number of anthraquinone compounds with high relative abundance in the two drying processes, obtained in release tests from the microcapsules.
The microcapsules prepared with the two carrier agents show the same relative abundances, indicating that no significant differences were found between the two encapsulants for the two drying processes.

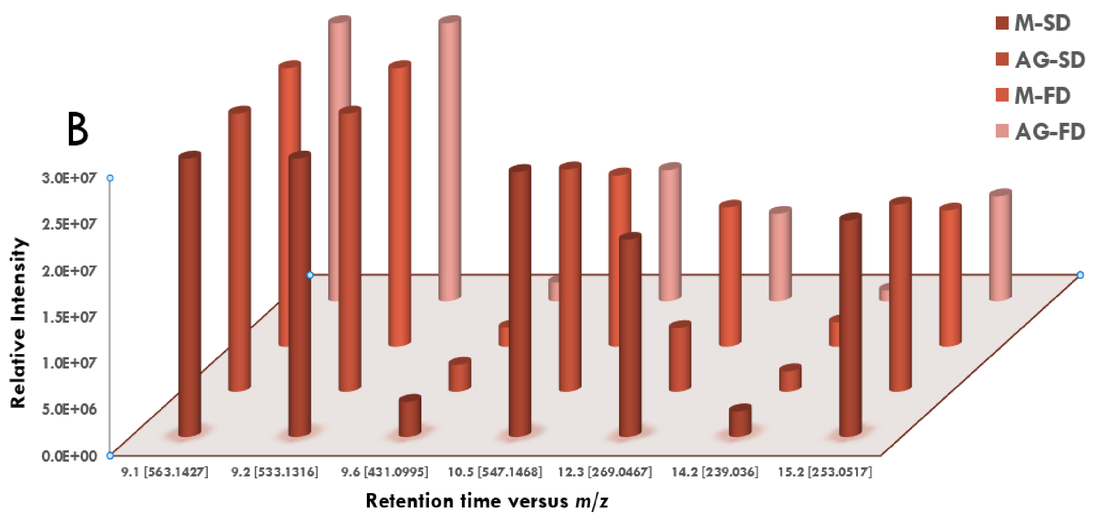


Fig. 4 Comparative diagram of the relative abundances acquired by LC-ESI(+/-)/HRMS of the major red chromophores present in sorghum (A) and rubea (B) extracts obtained, respectively, by freeze-drying encapsulated with maltodextrin (M-FD) and gum arabic (GA-FD), and by atomization encapsulated with maltodextrin (M-SD) and gum arabic (GA-SD).

Conclusions

Anthraquinone extracts encapsulated by lyophilization exhibited higher encapsulation efficiency (EE) and solubility.The chromatographic profile indicated that most of the dyes are glycosides, namely lucidin primeverosides, alizarin primeverosides, and rubiadine primeverosides2.In terms of pH stability, there were no significant differences between the two encapsulating agents, for the different values.In temperature stability, it was found that the two encapsulating agents offered protection for anthraquinones, with the freeze-drying process being the best. Encapsulation is a promising process, as it improves the stability of natural dyes, as it allows them to be obtained in powder form, making them easier to use, transport and storage.

References:
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