Microencapsulation of Thyme Oil by Coacervation: Production, Characterization and Release Evaluation

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INTRODUCTION AND OBJECTIVE

Nowadays, core-shell microcapsules have been investigated extensively for utilization in controlled release systems, especially in drug delivery, where the polymeric wall is a permeable element with porosity that can determine the release behaviour of core materials (Fairhurst *et al.*, 2008; Peña *et al.*, 2009; Romero-Cano *et al.*, 2002).

The controlled release systems are used to deliver compounds such us fragrances or flavours at prescribed rates, together with improved efficacy, safety and convenience. The objective of this work is to develop a coacervation process to produce polylactide (PLA) microcapsules containing thyme essential oil, having in view cosmetic applications. Generally PLA has been used for the microencapsulation of hidrosoluble active principles, but not with oils. The novelty of our process consists on dissolving PLA in dimethylformamide (DMF) which is a good solvent for PLA but in addition has high solubility in water. Upon contact with water, the homogeneous solution of PLA in DMF, promotes the precipitation of PLA around the thyme oil core (Martins *et al.*, 2009).
Chemical and structural characterization will be performed in order to understand microcapsule's properties and behavior. Release of the encapsulated essential oil will be also studied by developing a theoretical model for its diffusion across the PLA capsule and by performing experimental assays.

MATERIALS AND METHODS

Microcapsules of PLA with thyme oil were prepared according to the procedure described in Figure 1.

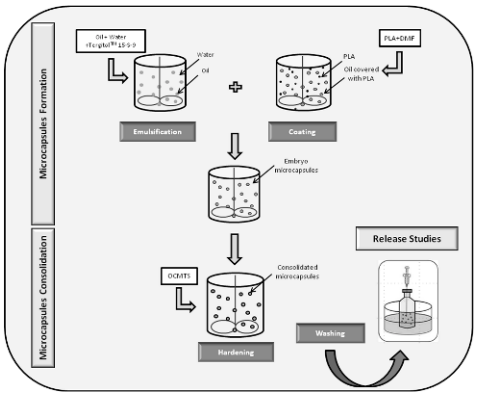

Figure 1: Process steps for microencapsulation of thyme oil by coacervation technique and for release studies.

PLA microcapsules can be described as spheres with a single-wall layer comprising inner (r_c) and outer radius (r_p) ($r_c < r_p$), which are assumed unchanged over the time. A schematic representation is presented on Figure 2. This assumption does not consider possible volume changes due to polymer degradation or swelling effects in the capsule.

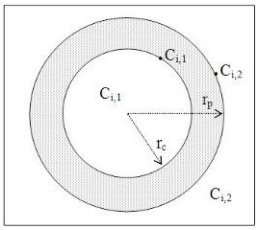

Figure 2: Schematic representation of a microencapsulated particle (Martins *et al.*, 2011).

The type of microcapsules considered in this study can be described as liquid core (essential oil of thyme) coated with a permeable membrane (polymer).

RESULTS AND DISCUSSION

Optical microscopy images of the microcapsules are shown in Figure 3. Figure shows that the droplets of thyme oil have been individually encapsulated as spherical particles with size distribution consistent with a bimodal distribution, and one can notice also the absence of agglomerates.

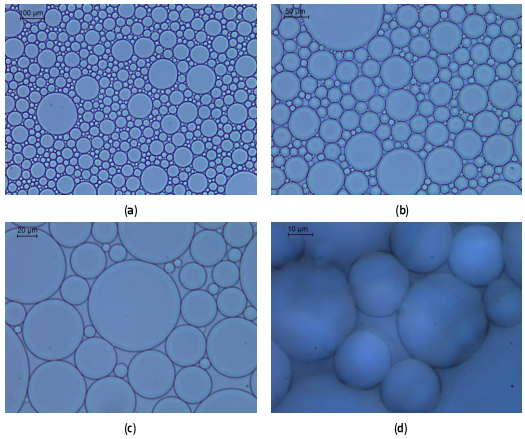

Figure 3: Optical microscopy of PLA microcapsules solution after the production and without washing, using Tergitol™ 15-S-9 as surfactant. Magnification of images: (a)100x; (b)200x; (c)400x and (d)1000x.

Figure 4 shows the experimentally measured particle size distributions, both in volume and in number, for

PLA microcapsules prepared with four different kinds of surfactants.

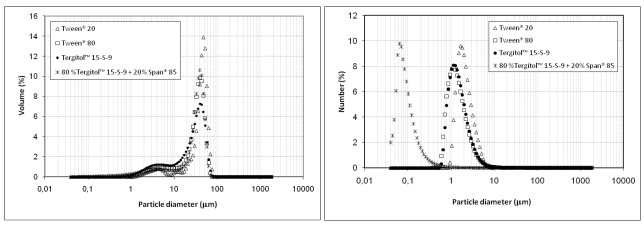

Figure 4: Particle size distribution of polylactide microcapsules with thyme oil for different surfactant systems and after washing the microcapsules. Distribution in volume (i) and in number (ii) (Martins *et al.*, 2010).

The distributions in volume for all the studied formulations have showed a similar distribution model, i.e., a bimodal distribution and pointed out that the use of Tergitol™ 15-S-9 generates smaller particles. The corresponding distributions in number were quite narrow and unimodal in shape.

Thymol and *p*-cymene were chosen as representative of the polar and nonpolar components of thyme oil, respectively. Figure 5 shows the release kinetics for thymol during the first hour of release.

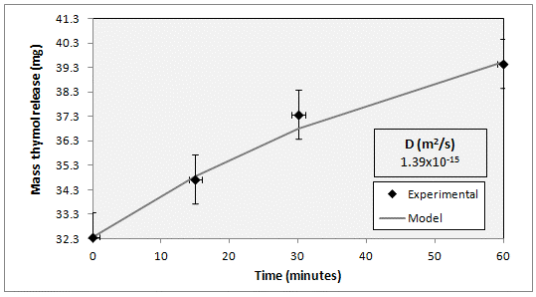

Figure 5: Comparison between experimental and model results for thymol released from PLA microcapsules solution for in first hour.

It was observed through Figure 5 that the diffusion coefficient was $1.39 \times 10^{-15} m^2/s$ for thymol, the polar component. For the apolar component, *p*-cymene, the diffusion coefficient for the first hour of release was 5.21×10^{-17} m^2/s, which is lower than that obtained for thymol. These differences can be ascribed to the distinct lipophilic solubility of the analysed thyme oil components and the obtained rather small diffusion coefficient values interpreted in terms of the very dense polymer matrix, which might constitute a significant hindrance effect.

CONCLUSIONS

In this work a novel coacervation technique for the microencapsulation of thyme oil with a biodegradable polymer (polylactide - PLA) was developed. The effect of using different surfactants systems in the particle size distribution, morphology and thyme oil encapsulation yield was investigated. It was concluded that when using Tergitol™ 15-S-9, a surfactant with a HLB of 13, an encapsulation yield of around 65% was obtained. Analysis by optical microscopy confirmed the spherical shape for all the produced microcapsules plus two predominant sizes, compatible with a bimodal distribution. The particle size analysis showed a bimodal distribution in volume with mean particle size around 30 µm using Tergitol™ 15-S-9. The release rate of thyme oil through the PLA microcapsules wall was evaluated and thymol and *p*-cymene chosen as representative of its polar and nonpolar components, respectively. It could be concluded that the release might be explained by a diffusion mechanism. The developed model was found to be in good agreement with the experimental measurements. The developed diffusion model could be extended to other single-layer microcapsule systems.

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ACKNOWLEDGMENTS

Financial support for this work was provided by LSRE financing by FEDER/POCI/2010, for which the authors are thankful and Isabel Martins acknowledges her Ph.D schorlarship by Fundação para a Ciência e a Tecnologia (FCT) (SFRH/BD/43215/2008).