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New microcapsules based on isosorbide for cosmetotextile: Preparation and characterization

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ABSTRACT

The preparation of polyurethane bio-based microcapsules containing the neroline fragrance aimed at cosmetotextile applications was investigated. The polyurethane shell material was synthesized by interfacial polycondensation using isosorbide and hexane diisocyanate as monomers.

Chemical characterization by means of IR-ATR and TGA confirmed the formation of a polyurethane wall and the fragrance encapsulation up to 73% of the dry microcapsules. The physicochemical characterization of neroline-loaded microcapsules was carried out, including thermal properties, size distribution, morphology and zeta potential. The spherical microcapsules of size ranging between 2 μm and 100 μm had a rough external surface. Their thermal stability up to 260 °C was favorable regarding their application to deposition onto textile fibers. According to their IsoElectrical Point of 5.38, these microparticles were anionic at neutral pH. An isosorbide-based cationic surfactant has been adsorbed at the surface of the microcapsules for charge reversal into cationic and stronger binding to negatively charged cotton fabric.

Besides, neroline-loaded microcapsules were fixed on pure cotton fabric by an impregnation technique. The reality and durability of the textile treatment were assessed by SEM and Gas Chromatography analysis. The cotton knitted fabric treated with fragrant microcapsules progressively released its microcapsule content; significant amount of residual neroline remained until 40 washing cycles.

1. Introduction

Microencapsulation technology is a growing area where microcapsules act as small containers of active materials to be released from their inner core under controlled conditions for specific purposes (Benita, 2005; Giamberini et al., 2015; Rodrigues et al., 2009). Thereby, this technology provides improved performance and increased durability of encapsulated materials in various application fields (Cheng et al., 2008; Martins et al., 2014). Hence, microencapsulation is used to obtain products with high added technical value (Benita, 2005; Casanova and Santos, 2016; Martins et al., 2014; Teixeira et al., 2012a).

Textile industry has also recently implemented novel technologies for applications in various fields, especially the cosmetic one (Persico and Carfagna, 2013; Teixeira et al., 2012b). The concept of smart textile has been introduced and several commercial cosmetotextile products are currently available in the market. On contact with human body, these products are designed to transfer to the skin an active material for cosmetic purposes (Persico and Carfagna, 2013). The majority of currently commercialized microcapsules are elaborated from thermosetting aminoplast resins which are prone to several drawbacks. In particular, they may contain residual formaldehyde that is a human carcinogen product and their degradation released formaldehyde. In addition, aminoplast resin is derived from petro-chemistry, which is a non-renewable resource (Rodrigues et al., 2008).

A current trend is going beyond by producing cosmetic loaded-microparticles from renewable resources, leading to green and bio-sourced materials (Azizi et al., 2014; Delebecq et al., 2012). This is expressed by the tremendous increase of the number of publications on bio-based polymers as specified by both ISI Web of Sciences and Thomas Innovations (Babu et al., 2013).

The first generation of such new polymers focused on deriving polymers from agricultural feedstocks like corn, potatoes, and other

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carbohydrate materials that open new high-value-added markets to agriculture. 1,4:3,6-dianhydrohexitols are examples of such bio-based carbohydrate chemicals. Interestingly, Isosorbide (1,4:3,6-dianhydro-D-sorbitol) attracts increasing attention, because it is inexpensive and available as large quantities (Babu et al., 2013; Feng et al., 2011; Fenouillot et al., 2010).

Accordingly, Azizi et al. (2014), synthesized isosorbide-based polyurethane microcapsules for cosmetotextile applications using interfacial polycondensation of isosorbide and 4,4′-methylene-bis(phenyl isocyanate) (MDI). The neroline fragrance (2-ethoxynaphtalene) was encapsulated inside microcapsules as a model case study. This first approach is currently extended by using 1,6-hexane diisocyanate (HDI) as an alternative to MDI with the expectation of bringing improved performances given in the following.

Both MDI-based and HDI-based polymers are semi-crystalline materials at room temperature, where the amorphous parts are glassy. Indeed the melting point and glass transition temperature (T_G) of MDI-isosorbide polyurethane are 187 °C and 235 °C respectively and those of HDI-isosorbide polyurethane are 110 °C and 190 °C respectively (Fenouillot et al., 2010). The soft segment of HDI yields a lower T_G than MDI; but both of them have quite high T_G, making such polymers quite thermostable materials. The substitution of HMI for MDI does not change the case so much with that respect.

- The first expected difference comes from solubility of fragrance in the polyurethane wall. Encapsulated fragrance can act as a plasticizer of the microcapsule walls and cause higher permeabity as T_G is lowered below room temperature. Owing to the aromatic nature of MDI, an aromatic fragrance such as neroline can swell the polymeric membrane, causing quite high permeability for fragrance release. Membrane materials prepared from the aliphatic MDI might show a better resistance to swelling by fragrance and show up higher encapsulation and slower sustained release of fragrance.
- A second improvement of substitution of HDI for MDI is the higher biodegradability of the aliphatic *n*-hexamethylene segment compared to the aromatic methylene-diphenyl segment. The hydrolysis products of HDI-isosorbide polyurethane are 1,6-hexanediamine and isosorbide whereas MDI-isosorbide polyurethane releases the aromatic diamine 4,4′-methylene bis(phenylamine).
- Another improvement with respect to the previous materials of Azizi et al. (2014) is the use of toluene in place of cyclohexane as the solvent of the oil phase in the emulsion used for interfacial polycondensation. Cyclohexane is a worse solvent of most polymer materials than toluene as inferred from their Hildebrand solubility parameters. Indeed, the Hildebrand solubility parameter of toluene is $\delta = 18.2$ MPa$^{1/2}$ whereas that of cyclohexane $\delta = 16.8$ MPa$^{1/2}$ is lower and much more different of the solubility parameter of most polymer materials (Hansen, 2000). For being more specific, the distances between polymer and solvents in the 3D-space of solubility parameter components for dispersion, polar and hydrogen bond interactions (δ_d, δ_p, δ_{hb}) introduced by Hansen (2000) is considered. The different components of δ were estimated from the increments per groups given by Hoftyzer and van Krevelen (van Krevelen and te Nijenhuis, 2009) and the distance between polymer and solvent was calculated as the root mean square of the differences of components: $\Delta\delta = [(\delta_d,\text{pol} - \delta_{d,\text{solv}})^2 + [(\delta_p,\text{pol} - \delta_{p,\text{solv}})^2 + [(\delta_{hb},\text{pol} - \delta_{hb,\text{solv}})^2]^{1/2}$ (Table 1). $\Delta\delta$ from cyclohexane to polyurethanes is much larger than from toluene to polyurethanes, showing that cyclohexane is a worse solvent than toluene. As it is considered that a polymer is “insoluble” in a solvent when $\Delta\delta > 5$ MPa$^{1/2}$, both solvent cause the polyurethane to precipitate at the oil-water interface, thereby creating a microcapsule. However an early precipitation caused by a very poor solvent (cyclohexane) leads to the formation of a thin polymer layer that blocks further polycondensation between the oil-soluble diisocyanate and the water-soluble isosorbide. Using toluene should allow easier polycondensation yielding polymers of higher molar masses, thicker wall and higher mechanical resistance of the microcapsules once the solvent has been evaporated. The same calculation done for neroline shows that neroline is a better solvent for MDI-PU than HDI-PU, as already qualitatively inferred above considering the plasticizer effect of neroline.
- A last change with respect to previous work by Azizi et al. (2014) is the choice of the fabric. Cotton fabric has been taken instead of polyamide because the hydrophilic cotton surface makes adhesion of hydrophobic microcapsules difficult to achieve; cotton fabric is a more severe test for the ability of microcapsule to bind to it. A specific surfactant based on isosorbide has been previously developed (Ben Abdelkader et al., 2016). As it is based on isosorbide, it is expected that it can bind favorably to the isosorbide-based wall of the microcapsules. It has been used for improving adhesion to negatively charged hydrophilic surfaces such as cotton. A stronger adhesion of microcapsules on fibers should yield a larger amount of microcapsules bound to the fabric.
- According to the above concepts, the present work aims at the development of polyurethane bio-based microparticles made of neroline fragrance encapsulated into polyurethane shell by means of interfacial polycondensation of 1,4:3,6-dianhydro-D-sorbitol (DAS) and 1,6-hexane diisocyanate (HDI).

Table 1
Solubility parameter components of the polymers and solvents, and distances between them in the (δ_d, δ_p, δ_{hb}) space.

δ (MPa$^{1/2}$)	MDI-PU	HDI-PU	Neroline	Toluene	Cyclohexane
δ_d	18.65	15.84	19.45	18.0	16.8
δ_p	3.33	3.59	2.49	1.4	0
δ_{hb}	9.56	9.99	4.32	2.0	0.2
Total δ	21.22	19.07	20.08	18.16	16.80
Rms distance between solubility parameter components $\Delta\delta$ (MPa$^{1/2}$)					
MDI-PU - solvent			5.4	7.8	10.1
HDI-PU - solvent			6.8	8.6	10.5

The polymerization reaction leading to the formation of the polyurethane shell was checked by FTIR-ATR analyses. Complementary morphological, thermal and interfacial characterizations of synthesized microcapsules were performed. Subsequently, fragrant microcapsules were applied to cotton fabric together with the isosorbide-based cationic surfactant and characterized for their morphology and fragrance release properties.

2. Experimental

2.1. Materials and chemicals

For the preparation of microcapsules, following chemicals were used as received: 1,6-hexane diisocyanate (HDI, Fluka 98%) and isosorbide (DAS, Acros 98%) as monomers, Polysorbate 80 (Tween™ 80, Aldrich 98%) and dibutyltindilaurate (SnDBDL, Aldrich 95%) respectively as emulsifier and catalyst. 2-Ethoxynaphthalene (neroline), employed as core material, was obtained by *O*-ethylation of β-naphthol with ethyl bromide (Azizi et al., 2011). Toluene (Sigma-Aldrich, 99.8%) was used, without further purification, as the dispersed phase of the oil-in-water emulsion. Emulsion was prepared using an Ultra-Turrax® T25 Basic homogenizer equipped with a S25 N 25 F shaft (IKA, Germany). Microencapsulation process was carried out in a thermostated double jacketed glass reactor (Sovirel, 1 L), equipped with a half-moon blade mixer, a mechanical digital control of stirring rate and an oil thermostat bath.

For the textile application of microcapsules, a knitted fabric (jersey, 100% cotton) bleached and mercerized, with a mass of 138 g·m^{-2}, was used. It was supplied by Esprit Maille (5015 Bouhjar, Tunisia). A polyurethane cross-linking agent (Politex SW/N) supplied by

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