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Controlled release of encapsulated methylene blue in a multilayered textile coating

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SUMMARY

We studied the formation of multilayered coating incorporating a β-cyclodextrin polyelectrolyte onto a pre-treated polyethylene terephthalate (PET) textile in order to obtain reservoir and sustained release properties towards bioactive molecules. This paper describes the alternate deposition by *dip-coating* onto the textile of chitosan (CHT) and a β-cyclodextrin polyelectrolyte (polyCTR-βCD) according to the layer-by-layer (LbL) principle. Textiles covered with up to 12 layers were characterized by gravimetry, infrared, zetametry. The building of the multilayer system was then achieved including methylene blue (MB) as bioactive model compound, complexed with polyCTR-βCD, and a release study of BM was investigated in batch. The results showed that the release profile of BM could be controlled by the number of layers in the system.

INTRODUCTION

In the last two decades, surface treatments of biomaterials have been investigated aiming to modify their physicochemical properties to control cell proliferation or adhesion, or to create drug delivery system (García-González *et al.* 2011). In this case, the main challenges are firstly to chemically modify such inert materials with soft methods in order to keep their original properties and secondly to adsorb a sufficient therapeutic dose of the drug that should then be released covering the critical healing period.

Cyclodextrins (CDs) and their polymers have proved to be the appropriate candidates for this purpose (Uekama *et al.* 1998). Due to the hydrophobic character of their cavities, they are known to form reversible inclusion complexes with many hydrophobic bio-active molecules and promote their solubility. So, CDs are widely used for their encapsulation properties as drug carriers (Li *et al.* 2004, Sun *et al.* 2012).

The aim of the present study is to build an innovative system of a multilayer assembly on a pre-treated PET using the LbL process, using a polymer of β-cyclodextrin (βCD) as a polyanion with an active molecule encapsulated in the cavities, and chitosan as a polycation, a biosourced biocompatible polymer. The LbL technique, developed by Decher over the past several years (Decher *et al.* 1992), is fast, easy to use, with a soft and biocompatible process (mainly in water) and is adjustable to several substrates.

This system was physically, chemically and biologically tested as drug delivery system.

EXPERIMENTAL PART

Non-woven PET, (65 g/m^2) was pre-treated by a high cross-linked copolymer of βCD and citric acid bearing anionic groups onto the surface (Martin *et al.* 2013). The multilayer assembly was then built by the dip-coating method (Decher *et al.* 1992). Pre-treated samples were alternatively dipped into a chitosan polycation solution, and a polyanion cyclodextrin polymer (poly-βCD, Fig. 1). Up to 12 layers were deposited. The same process was used, incorporating methylene blue as drug model (MB) in the system. The building of the multilayer assembly was monitored by Optical Waveguide Lightmode Spectroscopy (OWLS) and characterized by zetametry, microscopy (SEM) and physicochemical methods (FTIR, TGA, gravimetry). Biocompatibility was proved by culture in vitro of L132 cells. Release of MB was followed in distilled water and PBS solution by UV-visible spectroscopy.

RESULTS AND DISCUSSION

OWLS, zetametry and gravimetry confirmed that chitosan and poly-βCD were alternatively deposited on the surface. MB slightly disturbed the multilayer building due to its cationic character (Fig. 2).

SEM showed a significant partial filling of the gaps due to the fibers coating. The release study of MB showed a relation between the number of layers and the amount of released MB in the supernatant (Fig. 3). The layer by layer assembly allowed the sustained release of MB as no burst effect was observed. On the contrary, MB release occurred with a regular rate over 3 months.

Proliferation tests with L132 cells on samples coated with up to 12 layers proved their biocompatibility.

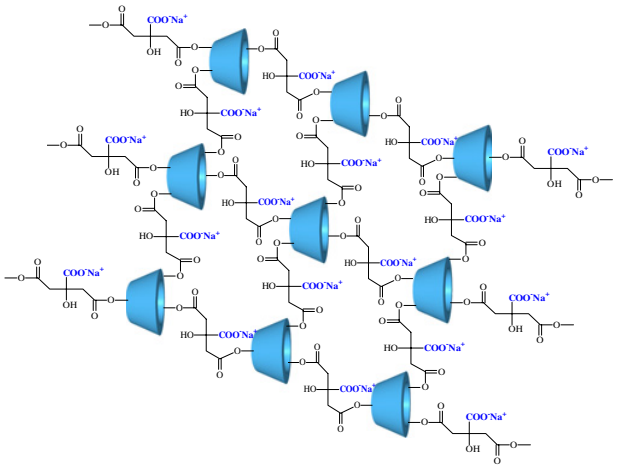

Figure 1. Structure of polyCTR-βCD.

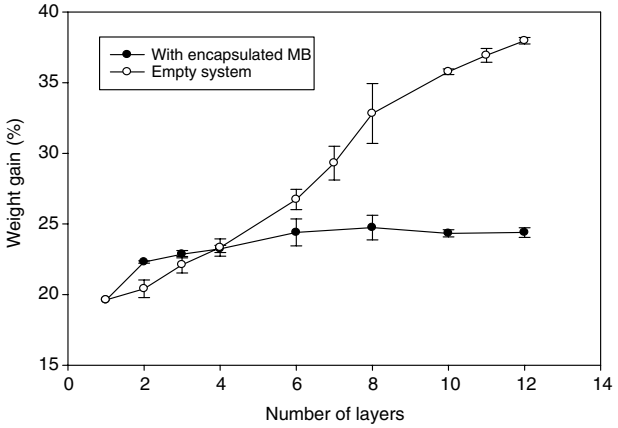

Figure 2. Comparative evolution of the cumulative mass on assemblies with or without MB.

CONCLUSION

We realized the controlled deposition of chitosan and cyclodextrin polyelectrolytes onto a textile support. The

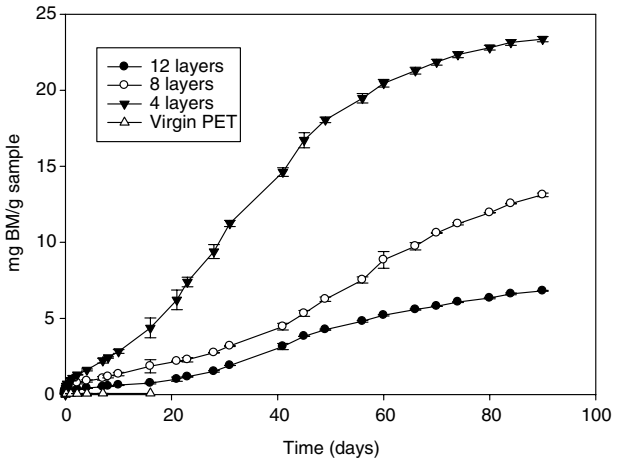

Figure 3. Release study of MB in multilayers assemblies in water at 37 °C under stirring.

L-*b*-L system allowed the control of the dose and the sustained release of MB. The biocompatibility of the system will allow us to investigate clinical applications, such as wound-healings, where antibacterial and healing properties of both used polyelectrolytes could offer a relevant added-value.

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