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Influence of pH upon *in vitro* sustained dye-release from oil-core nanocapsules with multilayer shells

Urszula Bazylińska[a], Piotr Warszyński[b], Kazimiera A. Wilk[a,*]

[a] *Faculty of Chemistry, Wroclaw University of Technology, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland*
[b] *Institute of Catalysis and Surface Chemistry Polish Academy of Sciences, Niezapominajek 8, 30-239 Kraków, Poland*

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ABSTRACT

An ideal drug-delivery system should combine both the stimuli-responsiveness and biocompatibility to enhance the drug bioactivity as well as to reduce side effects. In the present work we focused on the influence of pH upon *in vitro* sustained dye-release from multilayer oil-core nanocapsules. Three types of polyelectrolytes (PEs), two strong synthetic, poly(diallyldimethylammonium chloride) (PDADMAC) and poly(sodium 4-styrenesulfonate) (PSS) and one weak biocompatible: poly(L-lysine) hydrobromide, were used for the formation of sustained release, oil-core nanocapsules via layer-by-layer (LbL) self assembly approach. All nanocarriers coated with PSS/PLL or PSS/PDADMAC bilayers were created on the oleic acid nanoemulsion templates stabilized by dicephalic-type surfactant and loaded with hydrophobic, cyanine-type photosensitizer, IR-786. Nanocapsules with different thicknesses of the PE shell and average size around 100 nm, demonstrated good capacity for cyanine IR-786 encapsulation. We determined the sustained release of cyanine dye in different pH-conditions – physiological, acidic and alkaline. The *in vitro* release profiles were obtained spectrophotometrically and interpreted in terms of diffusion-controlled processes. They proved that pH condition had some influence on the release rate of the dye. It indicates on the necessity of appropriate selection of anionic and cationic polyelectrolytes in the fabrication process of pH-responsive and long sustained release nanocapsules.

1. Introduction

Biomaterials are currently receiving significant attention because of their enormous impact in nanomedicine and biopharmaceutical industry. Since it is evident that for any clinical application biocompatibility of the materials is crucial, many efforts to create pure and highly biocompatible polymers have been undertaken. It has allowed scientists to apply them in various areas including tissue engineering, wound dressings and drug delivery [1–3]. In the recent years the question of drug encapsulation at the nanoscale level has also raised much interest in part due to the advancement and optimization of the biomaterials that is used to develop the nanocapsules [4,5]. The polymeric nanocapsule shells are commonly composed of biocompatible and biodegradable polymers, while their core generally consists of an oil phase with a dissolved hydrophobic drug [5,6]. Those polymeric nanocarriers have many advantages over conventional drug delivery systems. They can increase the bioavailability, improve solubility and prolong the shelf-life of many potential drugs, which are otherwise difficult to deliver. Furthermore, oil-cored nanocapsules can also provide ingenious treatment by improving targeted delivery and controlled release, reducing the drug dosage frequency and increasing the patient compliance [7]. The fundamental issue for nanocapsules biocompatibility is evaluating their potential cytotoxicity, either because of their size and shape or because of the interaction of the nanocarrier surface with target cells. Therefore, the key approach to form optimal delivery nanosystems (both stimuli-responsive and biocompatible ones) consists in using the capacity of some biocompatible polyelectrolytes (PE) to change their macromolecular characteristics in response to external stimuli such as pH, temperature, ionic strength change or illumination [1,7,8].

The layer-by-layer (LbL) self-assembly of pH-sensitive building blocks is a promising approach to fabricate new biomaterials with well-defined architectures and tunable properties [9,10]. Due to their potential applications as stimuli-responsive nanocarriers of pharmaceuticals for controlled drug release and the ability to overcome the multiple biological barriers, there is a considerable interest in this fascinating class of materials [11–13]. The recent extension of the LbL technique for encapsulation of drug-loaded liquid cores, *i.e.*, oil-in-water nanoemulsions, has led to a viable alternative for preparing capsules with multilayer shells with high loading capacity and controlled size and permeability [5,14,15]. Moreover, the use of appropriate pH-dependent

* Corresponding author. Tel.: +48 71 320 28 28; fax: +48 71 320 36 78.
E-mail address: kazimiera.wilk@pwr.wroc.pl (K.A. Wilk).

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