



Pore size effect on the formation of polymer nanotubular structures within nanoporous templates



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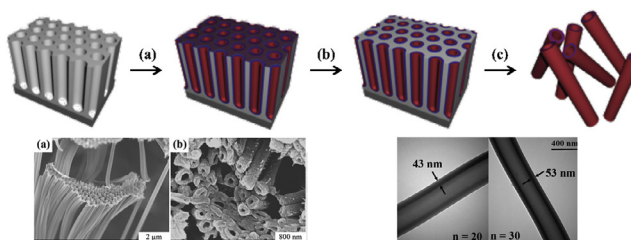
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HIGHLIGHTS

- Cylindrical nanoporous templates with various pore diameters were synthesized.
- Polymer nanotubes were obtained based on the template-assisted surface modification by nano-films.
- Wall thickness of the nanotubes was affected by the pore size of porous templates.

GRAPHICAL ABSTRACT



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ABSTRACT

Nanotubular structures have recently received much attention due to their potential applications in biosensors, drug delivery systems, electronic devices, and many others. The layer-by-layer (LbL) deposition technique on 3-dimensional templates has been one of the most popular methods for the formation of nanotubular structures. Any size, shape, and composition template can be utilized and the desired amount of various materials can be readily incorporated within the thin film geometry with nanoscale control. Therefore, the morphological features of those materials can easily be tuned by varying the structural properties of templates. However, LbL deposition within a confined geometry actually shows somewhat different results due to the geometrical restriction, which is still not fully understood so far. In the present study, in order to investigate such a template effect on the LbL process, we utilized a cylindrical nanoporous anodic aluminum oxide (AAO) structure as a template for the LbL process, yielding polymer nanotube structures. By varying the pore size of the porous templates, we determined the LbL process within porous structures was strongly governed by the geometrical characteristics of the utilized templates.

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1. Introduction

Recently, nanotube structures incorporating a variety of functional materials have received considerable attention as one of the most versatile and important materials for broad ranges of applications ranging from electronic devices to drug delivery

systems [1–4]. Among various preparation methods to realize such nanotube structures, the layer-by-layer (LbL) deposition on 3-dimensional porous templates has been regarded as one of the most powerful and popular methods for the creation of nanotube structures because the typical advantages of the LbL deposition method on 2-dimensional flat substrates yielding multilayered thin films can also be easily extended to 3-dimensional porous substrates, leading to nanotube structures [5–13]. The versatility of the LbL method has allowed a wide range of materials to be assembled on the basis of complementary interactions including polymers, nanoparticles, graphene, proteins, and DNA [14–18]. The amount of

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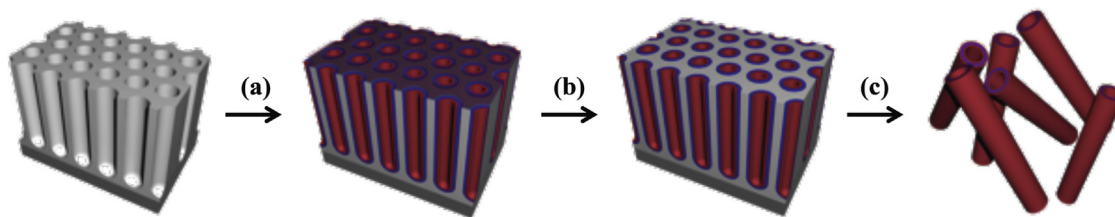


Fig. 1. A schematic depiction of the preparation of polymer nanotubes realized within AAO templates by the layer-by-layer deposition method: (a) alternative LbL deposition of polyanionic and polycationic electrolytes within the AAO templates; (b) selective removal of multilayer films formed onto the AAO templates; (c) dissolution of the AAO template by either acid or base solution.

incorporated materials within the thin film geometry and the thickness of the multilayer film can be readily controlled with nanoscale control by the number of sequential adsorption processes until the desired number of bilayers is attained. Furthermore, templates of various sizes, shapes, and compositions can be utilized, allowing easy adjustment of the morphological features of the created nanotube structures. Therefore, many researchers have utilized the template-assisted LbL deposition method based on the assumption that the thin film grows homogeneously from the entire surface of 3-dimensional substrates including inner pore walls regardless of their structural properties, yielding an ideal conformal coating. However, several researchers recently demonstrated that the wall thickness of polymer nanotube structures prepared from a nanoporous template is somewhat different than that assembled on a flat substrate [6,7,19–22]. Such different multilayer growth behavior between different templates has not been completely understood. It prevents precise control over the structural properties of the resulting nanotubes, even though the thickness of

multilayer thin films representing the amount of incorporated molecules is one of key parameters to determine desired functionalities of materials prepared.

In this study, we successfully obtained polymer nanotube structures by the LbL deposition method on anodic aluminum oxide (AAO) membranes with various pore diameters containing hexagonally ordered cylindrical nanopores. It was clearly observed that multilayer growth within a narrow pore, representing the wall thickness of polymer nanotubes, is strongly governed by the pore size of the template used. Furthermore, such variation in multilayer thickness existed within a certain pore size window and is no longer shown when pore size is large enough, yielding similar or thicker growth behavior than that on a flat substrate. Thus, a critical pore size exists which directly affects molecular assembly within pores. One important result of our investigations is that the amount of adsorbed molecules within pores can possibly be controlled by changing the pore size of the porous templates, allowing precise control on desired functionalities. As a result, this approach can be

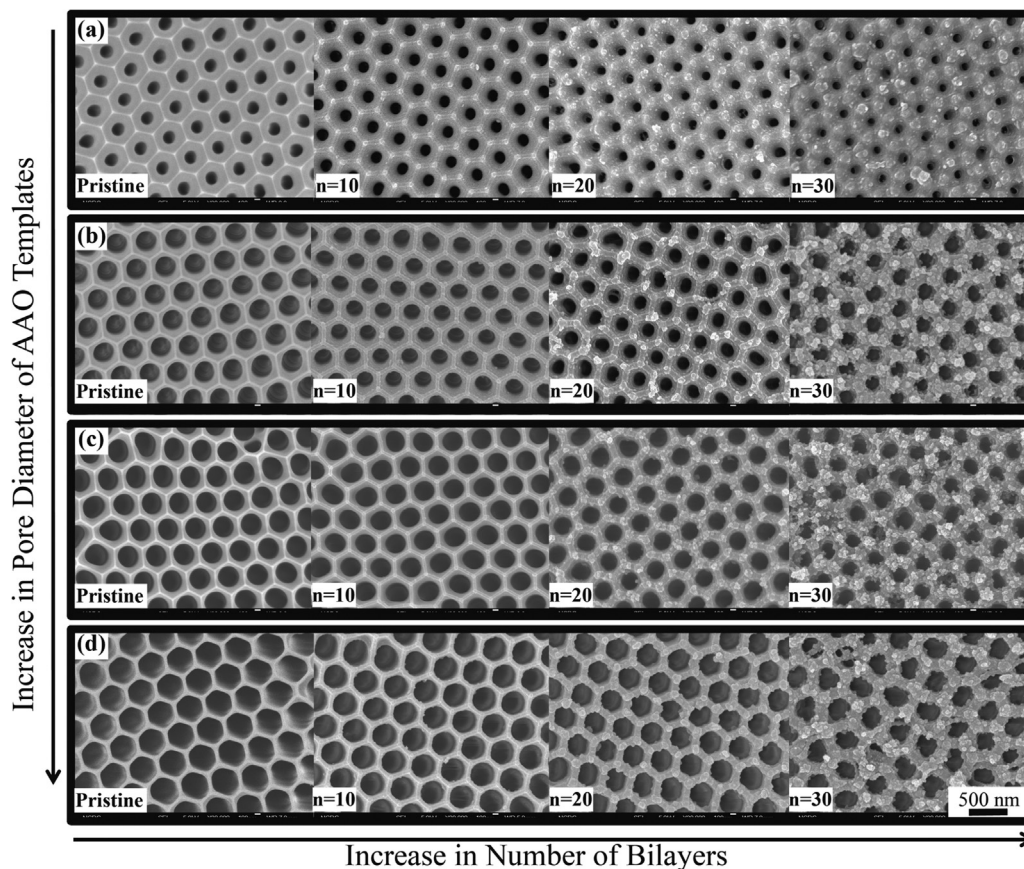


Fig. 2. FE-SEM images of $(\text{PSS}/\text{PAH})_n$ multilayer coated AAO templates with various pore diameters as a function of number of bilayers (n : number of bilayers): AAO templates with pore diameter of (a) 195, (b) 300, (c) 345, and (d) 430 nm.

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