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Multi-step functionalization procedure for fabrication of vertically aligned mesoporous silica thin films with metal-containing molecules localized at the pores bottom

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ABSTRACT

A novel method for fabrication of precisely functionalized nanomaterials was developed. Presented here multi-step functionalization procedure can be applied for vertically-aligned mesoporous silica thin films. As an example, we presented the syntheses of films, precisely functionalized by the propyl-copper-phosphonate and the propyl-silver-carbonate groups located exclusively at the pores bottom. Obtained functional materials were characterized by a few physical and chemical techniques: transmission electron microscopy, X-ray scattering, differential pulse voltammetry and infrared spectroscopy supported by the numerical simulations. On this base, we prove the efficiency of the synthesis method. We expect that presented multi-step functionalization procedure can be easily generalized and applied for fabrication of the novel porous silica-based functional materials.

1. Introduction

Functional materials based on the porous silica matrix and the transition metal-containing active groups have become very attractive architectures due to their unique properties [1]. They can be considered as the compounds possessing particular properties, precisely tailored to the specific functions [2]. These properties are usually a result of a purposely designed molecular structure and carefully elaborated synthesis route. The materials possessing such unique properties have found various applications in electronics [3,4], energy storage [5], sensing [6], catalysis [7,8], or even bioactive systems [9]. Especially, the thin films formed by the functional materials are crucial for the implementation in the nanoelectronic [10,11] or optoelectronic systems [12,13] as well as in solar cells [14,15].

The bottom-up approach to nanotechnology [1] has opened a promising way for the fabrication of materials having extremely ordered structure on the molecular level with means of self-assembly method. The properties of such materials are very sensitive to the supramolecular arrangement of the functional units, that determines the molecular polarity and hence all molecular properties [16]. The intermolecular interactions are responsible for cooperative phenomena and for the appearance of new features at the space charge transfer. Thus, the same molecule behaves substantially differently in a different environment or being distributed in a various way in the matrix. This feature can be applied for designing of materials for the use in optoelectronics. The most applicable form of such materials is a thin film. Nevertheless, designing and fabrication of functional thin films with desired features is still a challenging task. Moreover, the synthetic routes are often complicated and the structural characterization can be ambiguous. However, a possibility of the physical properties arrangement to achieve desired functionalities is still worth the effort. Having a proper matrix one can obtain the material tailored for a particular application. However, the *condicio* sine *qua non* in this case is fully controllable functional units distribution inside matrix volume. It is difficult but possible to achieve. As we shown in our previous works it is possible to fabricate a precisely functionalized silica-based strongly antimicrobial system, that is safer for the environment than commonly used disinfectant agents [17], or novel materials for optoelectronic: silica thin films possessing tunable nonlinear optical response [18]. In both cases, the key was extremely precise matrices functionalization

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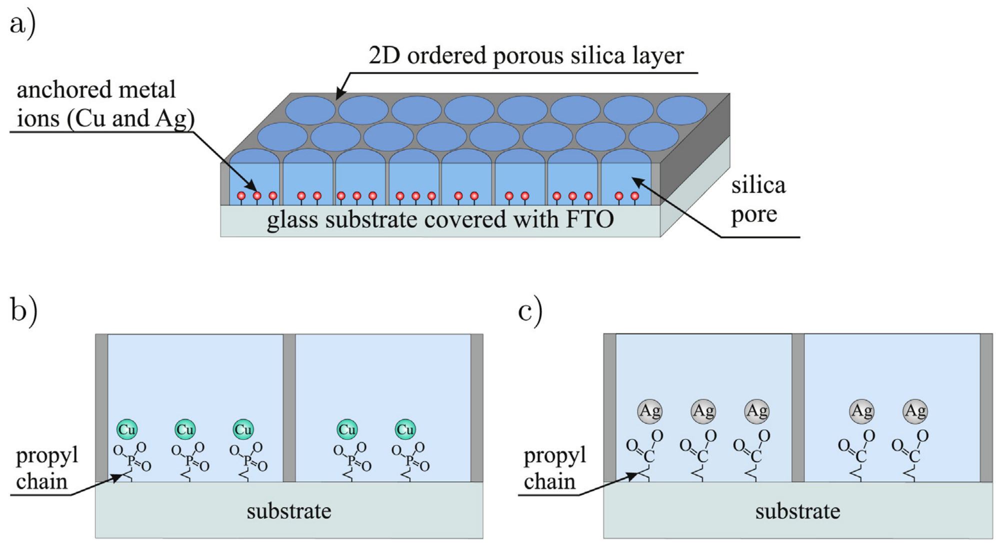

Fig. 1. Schematic representation of the ordered porous silica thin layers with vertically aligned pores, containing copper and silver ions bounded at the pores bottom. Prospective view (a) and zoom-in of the species containing copper (b) and silver (c).

and control of functional units distribution. All these results prompted us for searching for an even more accurate way for functionalization of nanostructured materials.

In the paper, we propose an innovative approach to a synthesis of completely novel thin film composite material based on the vertically aligned mesoporous silica matrix: ***the multi-step functionalization procedure***. This synthesis route is designed in such a way that each of silica channel contains the metal ions anchored exclusively at the pores bottom.

The method presented here provides an opportunity to achieve the precisely designed molecular structure of the samples. Moreover, this method can be easily generalized to be used for the fabrication of a new, very sophisticated nanomaterials, especially for the use in nanoelectronics devices. In the case of using derivatives of Mn_{12} molecular magnets, it is possible achieving of the regular layout of bistable magnetic units, that can play the role of a super-dense memory or a basic element of the molecular implementation of artificial neural networks (work in progress). However, in a first step, the

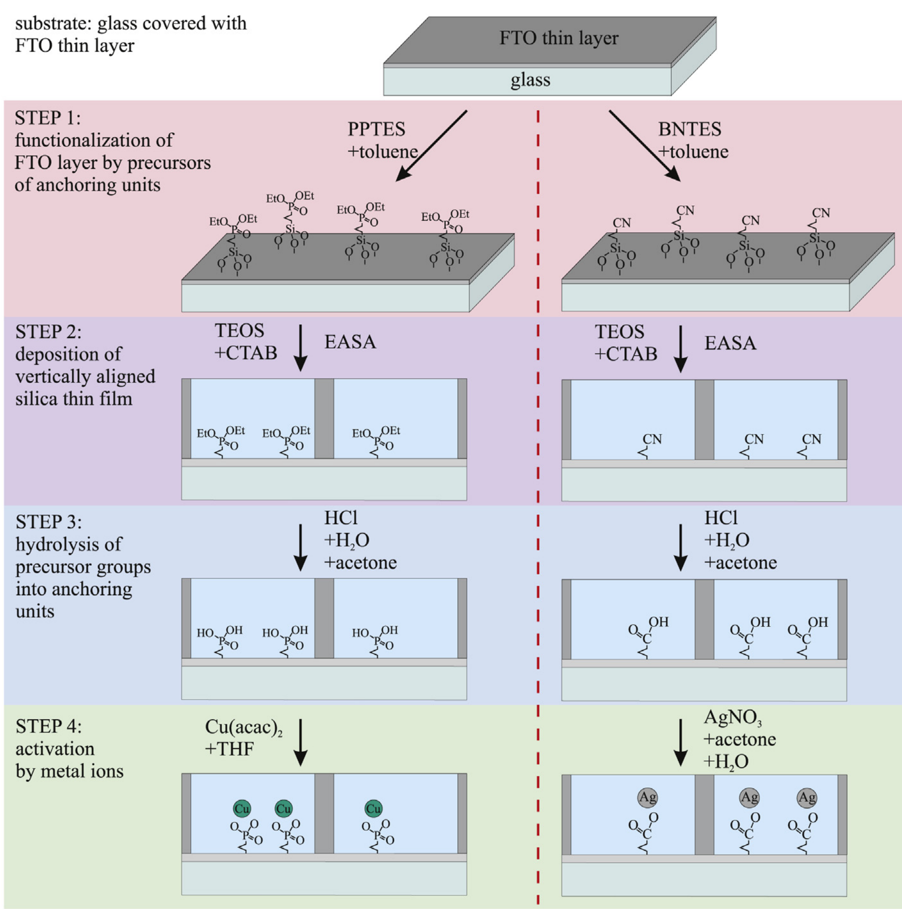

Fig. 2. Schematic representation of the synthesis procedure for fabrication of vertically aligned thin mesoporous silica layers containing metal ions at the bottom of channels. Denoting: PPTES - diethylphosphonatepropyl triethoxysilane, BNTES - butylonitrile triethoxysilane, TEOS - tetraethyl orthosilicate, EASA - electro-assisted self-assembly method, THF - tetrahydrofurane, Et - ethyl group.

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