Applied Surface Science 355 (2015) 290–297

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

# Fabrication of the replica templated from butterfly wing scales with complex light trapping structures

Zhiwu Han, Bo Li, Zhengzhi Mu, Meng Yang, Shichao Niu*, Junqiu Zhang, Luquan Ren

*Key Laboratory of Bionic Engineering (Ministry of Education, China), Jilin University, Changchun 130022, PR China*

ARTICLE INFO

*Article history:*
Received 13 March 2015
Received in revised form 4 July 2015
Accepted 17 July 2015
Available online 20 July 2015

*Keywords:*
Bio-template fabrication
Sol–gel
Selective etching
Butterfly wing scales
Light trapping structure
Bionic surface

ABSTRACT

The polydimethylsiloxane (PDMS) positive replica templated twice from the excellent light trapping surface of butterfly *Trogonoptera brookiana* wing scales was fabricated by a simple and promising route. The exact $SiO_2$ negative replica was fabricated by using a synthesis method combining a sol–gel process and subsequent selective etching. Afterwards, a vacuum-aided process was introduced to make PDMS gel fill into the $SiO_2$ negative replica, and the PDMS gel was solidified in an oven. Then, the $SiO_2$ negative replica was used as secondary template and the structures in its surface was transcribed onto the surface of PDMS. At last, the PDMS positive replica was obtained. After comparing the PDMS positive replica and the original bio-template in terms of morphology, dimensions and reflectance spectra and so on, it is evident that the excellent light trapping structures of butterfly wing scales were inherited by the PDMS positive replica faithfully. This bio-inspired route could facilitate the preparation of complex light trapping nanostructure surfaces without any assistance from other power-wasting and expensive nanofabrication technologies.

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## 1. Introduction

Over the past few years, there has been increasing interest in the research of fantastic surfaces triggered by nature, such as the anisotropy surface of the rice leaves [1,2], the self-cleaning surface of the lotus leaves [3–8], the antireflection surface of moth eyes [9–11], the fog collection system in cactus and the iridescence surface of the butterfly wings [12–14]. Inspired by these functional surfaces in biological systems, researches on replicating them into new functional materials are coming to the forefront of research in nanotechnology. With the help of various nanofabrication techniques, which are considered as the key factor in the realization of multifunction, materials scientists and engineers have made great efforts to study the preparations of nanostructure surfaces. According to previous reports, coating porous or multilayered films on the surfaces of devices and fabricating nanostructures directly on the surfaces of devices are two different kinds of available approaches to fabricating functional surfaces [15]. Jung et al. have fabricated the antireflective nanostructures (ARS) by etching the curved surface of polymer microlens with a metal annealed nanoisland mask [16]. Xu et al. have fabricated biomimetic hierarchical arrays based on the combination of self-assembled polymer spheres and nanoimprint lithography [17]. Tian et al. have fabricated colorful humidity sensitive photonic crystal (PC) hydrogel by infiltrating acrylamide solution into a P (St-MMA-AA) PC template and subsequently photo-polymerizing [18]. Xi et al. have reported the fabrication of $TiO_2$ and $SiO_2$ graded-index films deposited by oblique-angle deposition, which demonstrated potential for antireflection coatings [19]. In short, the nanofabrication of these natural structure surfaces has attracted more and more attention in various fields, especially photoelectrical device, optical sensor and photonic crystal [20–22].

Butterfly wings, as one of the most typical biological prototypes in the field of bionics, possess various remarkable properties, especially high-performance optics [23], such as photonic crystal structures (PCs) [24–26], structural color [27,28], light trapping effect [29,30] and so forth. Although the optical mechanism and structural characterizations have been well investigated for a long time, the research on bionic preparation of these subtle nanostructures in butterfly wing scales has been greatly restricted. On the other hand, the exact combination of the three dimensional (3D) structure and cuticle complex refractive index is beyond capabilities of the existing nanofabrication techniques. In spite of this, the potential valuable application prospect still inspired scientists and engineers to devote themselves to mimicking the distinctive

* Corresponding author at: Key Laboratory of Bionic Engineering (Ministry of Education, China), Jilin University, No. 5988, Renmin Street, Nanguan District, Changchun, PR China.
*E-mail address:* niushichao@jlu.edu.cn (S. Niu).

http://dx.doi.org/10.1016/j.apsusc.2015.07.119
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surface nanostructures of butterfly wings. The photonic structures of the colorful butterfly wing scales have been fabricated using soft lithography technique [31], low-temperature atomic layer deposition [32,33], colloidal self-assembly, sputtering and atomic layer deposition, even combining all these layer deposition techniques together [34]. Not only sorts of surface processing technology but also a variety of materials were employed to fabricate replicas of the multi-layered scales on the surface of butterfly wing, such as polyelectrolyte multilayer films [35], carbon nanotube [36], polypyrrole [37], oxidizing material include $SiO_2$ [38], $Bi_2WO_6$ [39], $Fe_3O_4$ [40], $SnO_2$ [41] and so on. In addition, some works were also focused on metallic replicas [42,43] and polymer replicas [44,45].

Although researchers have made a lot of efforts in this regard, there still desperately lack a straightforward technique to realize the fabrication of the multifunctional surfaces of butterfly wing scales, especially considering the cost and production efficiency. So, it is necessary to develop a high efficiency and low cost technique to realize the outstanding bionic functional surfaces of butterfly wing scales. Bio-template method, as a new method of fabricating multifunctional materials with bionic morphologies and structures, has attracted more and more attention of many scholars. In previous work, we had achieved an ingenious "negative" replica of the functional surface of butterfly wing scales using bio-template method [38,46]. However, it remains a challenge to fabricate the positive replica of the light trapping surface of butterfly wing scales.

In this paper, the bio-template method was improved and optimized. A straightforward, general and facile method was presented to fabricate the positive replica of *Trogonoptera brookiana* butterfly wings, which has excellent light trapping properties. In this work, we select the *Trogonoptera brookiana* butterfly as the bio-template and the excellent light trapping characteristics were characterized by reflectance spectrum. The structure and morphology of the wing scales were characterized by field emission scanning electronic microscopy (FESEM). In order to prepare the $SiO_2$ negative nanostructures replicas, a simple and high effective synthesis method combining a sol–gel process and subsequent selective etching was adopted. Based on the $SiO_2$ negative replicas, the polydimethylsiloxane (PDMS) positive replicas were successfully synthesized by a secondary bio-template method. At last, it was found that the PDMS positive replica really inherited the original light trapping structures of butterfly wings. Furthermore, this work would open up possibilities for extensive study of mimicking novel bio-inspired functional materials.

## 2. Materials and methods

### 2.1. Materials

Analytic grade reagents hydrochloric acid and tetraethyl orthosilicate (TEOS) were provided by Beijing Chemical Works. Ethanol absolute, diethyl ether, concentrated nitric acid, perchloric acid, and polydimethylsiloxane were provided by Tianjin Fine Chemical Co., Ltd.

The wings of butterfly *Trogonoptera brookiana* occurring in tropical rainforest at altitudes from 500 to 1500 m in Southeast Asia, possess light trapping property which was confirmed as reported in our previous work. Thus, its wing scales were selected as the biological prototype in this work. There are triangular and rhombus green zones distributing on the back of front and hind wing, respectively. Because of the brighter structural color, the triangular green areas of butterfly *Trogonoptera brookiana* front wing, which are covered by vivid green scales, were taken as the experimental materials to do further research.

Butterfly wing samples of uniform size (15 mm in length and 10 mm in width, rectangular) were cut off from the triangular green areas in a perpendicular and parallel direction to the ridge veins, respectively. In order to confirm that the butterfly wing samples were clean, some pre-processing was conducted. Firstly, each sample was soaked by aether for 10 min to removing proteins and fattiness on the samples surface. Afterwards, the dehydration treatment in ethanol absolute with duration time of 15 min for each specimen was taken. The purpose of conducting the dehydration pre-processing was to increase the mechanical strength and stability of the treated tissues. Then, the samples were dried naturally in air.

### 2.2. Spectrum characterization

The reflectance spectrum of *Trogonoptera brookiana* butterfly wing scales was measured using a miniature fiber optic spectrometer (Ocean Optics USB4000) equipped with a halogen tungsten lamp source (Ocean Optics LS-1-LL). The spot size of the incident beam on the wings surface was ~2 mm, and the wavelength of the reflectance spectrum varied in the range of 400–900 nm. What is more, the reflectance spectra of chitin, PDMS and PDMS replicas were obtained using the same method as well. The PDMS replicas were examined by fourier transform infrared spectroscopy (FTIR) measurements using a Bruker EQUINOX 55 instrument. X-ray diffraction (XRD) measurements of $SiO_2$ negative replicas were carried out with a Bruker-AXS X-ray diffractometer system by applying Cu K$\alpha$ radiation.

### 2.3. Preparation of the $SiO_2$ negative replica

Firstly, the butterfly wing samples were sandwiched between two glass slides, of which both ends were clamped by clips with proper force. Using a micropipette, a suitable amount of the sol–gel precursor solution, a reaction product of TEOS and hydrochloric acid (3:1 in volume), was added to the edge of the assembly with a volume of 4–6 μL. Then, the assembly was heated at 120 °C for 30 min in an electrothermal constant-temperature dry box to further solidify the precursor solution on the surface of the wing samples. Next, the whole assembly was dipped into a mixture of concentrated nitric acid and perchloric acid (1:1 in volume) while heating at 130 °C for 30 min to remove the original organisms. Finally, the whole assembly was washed by ultrasonic oscillation for 5 min in deionized water to get rid of the residue.

### 2.4. Preparation of the PDMS positive replica

The next step is the preparation of the PDMS positive replica. Firstly, the pre-polymer and the crosslinking agent were uniformly mixed and stirred (10:1 in weight) in a glass beaker to synthesize the PDMS. Then, the glass beaker which contained the PDMS was placed in a vacuum oven to evacuate the air bubbles. The PDMS was poured onto the $SiO_2$ negative replica, and we got the PDMS/$SiO_2$ negative replica assembly. After that, the PDMS/$SiO_2$ negative replica assembly was placed in a vacuum oven to evacuate the air bubbles again and heated at 80 °C for 120 min to complete heating curing process. After cooling at room temperature, we took the PDMS off the $SiO_2$ negative replica and got the PDMS positive replica.

### 2.5. Measurements

The 2D morphologies and structures of a single scale surface, the $SiO_2$ negative replica and the PDMS positive replica were obtained with the help of field emission scanning electronic microscopy (FESEM: JEOL JSM-6700F). These data would be used to analyze the inheritance accuracy of the $SiO_2$ negative replica and the

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