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ARTICLE TYPE

High light absorption properties and optical structures in butterfly *Heliophorus ila Lvcaenidae* wing scales

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When the sunlight irradiates on surface of butterfly wings, it could be absorbed by the microstructures in the wings scales and converted into heat to maintain butterfly's metabolism. This phenomenon is an inspiration uninterruptedly facilitating the scientific research in solar energy utilization. In this study, the absorption characteristics of seven species of butterfly were investigated by using spectrometer. It was found that the butterfly *Heliophorus ila Lvcaenidae* showed more efficient absorption capability (absorptivity was about at 85%) compared with other species in the wavelength span from 230 nm to 850 nm. Then, the morphology and structures of the butterfly *Heliophorus ila Lvcaenidae* wing scales were examined by Scanning Electron Microscope (SEM) and Transmission Electron Microscope (TEM). The results showed that there were two kinds of scales distributed on wing surface of the butterfly *Heliophorus ila Lvcaenidae*. Finally, the optical mechanisms were revealed by theories of multiple reflection and resonator. It was confirmed that the hierarchical hollow nano-architectures of the scales were responsible for the high-efficiency absorption behaviour. This study could be used as a theoretical reference for the subsequent bionic design of structural materials for solar energy utilization.

Introduction

As the typical hotspot of the bionics field recently, buttery wings are endowed with a diversity of excellent properties.[1] In recent years, scholars have done a lot of research on the species, warning colorations and scales microstructure.[2-4] The organized distribution of the scales can be observed under quite lower magnification. These scales are composed of a chitin materials.[5] Due to different optical effects, for example, anti-reflection, diffraction, light scattering as photonic crystal structure as well,[6-11] scales originate many sorts of colors which can be employed for signal transmission, mimicry, alerting and other purposes.[12-14] In terms of applications, bionic products originated from butterfly have been appearing in various occasions. For example, the polarization-sensitive characteristicinspired by butterfly structural colors can be used for anti-counterfeiting and decoration techniques.[15] By simulating scales optical characteristics in different spectrum, stealth skills[16-18] and thermal or gas sensor were developed.[19,20] Super black carbon film were manufactured by imitating scales microstructure.[21,22] Also, the butterflies scales have been adopted as templates for fabricating light trapping specimen[23,24] and other special function devices[25-28] by scholars all over the world.

Recent studies showed that there were certain species appearing in very early spring (April or May) at high-altitude mountain land, energy was used to protect their body from cold climate, which depended on the efficient sunlight ab-sorption of their wing surface scales (Fig. 1). In addition, during breeding season, more energy was needed to gestate eggs, while the energy couldn't be provided by their body since the declination of its mobility. Therefore, more solar energy should be obtained for energy demand during this period. The efficient absorption relied on not only the arrangement format, but also mainly the delicate nano-architecture inner scales.

In this work, a spectrometer was employed to test optical characteristics of 7 kinds of butterflies. The results illustrated that the butterfly *Heliophorus ila Lvcaenidae* scales had the best sunlight absorbing effect with absorptivity arriving at 85%. The microstructures of butterfly *Heliophorus ila Lvcaenidae* scales were studied with SEM and TEM and the simplified models were then built up. The cross section image of butterfly *Heliophorus ila Lvcaenidae* scales had two hierarchies microstuctures. The upper part had a hollow triangle form that resulted in multiple reflections and refractions. The bottom hollow layer formed a rectangular resonant cavity model which had light trapping effect. The solar egergy was absorbed efficiently for the integrated function of these two mechanisms. This study demonstrated an enlightening way to exploit bionic designning on structural materials of solar heat utilization.

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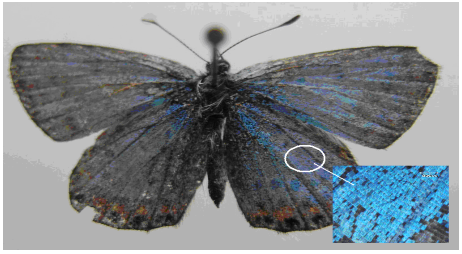

Fig. 1 Image of Butterfly *Heliophorus ila Lvcaenidae* and distribution of the scales on its wing surface.

1. Materials and Methods

1.1 Spectrometer

Seven species of butterfly were studied: *Papilio machaon*, *Parnassius stubbendorfii*, *Papilio maackii*, *Brenthis daphne*, *Apatura ilia*, *Heliophorus ila Lvcaenidae* and *Troides brookiana*. Test specimens were taken from single pure color areas of abdomen wings and with complete scales to avoid influence on the optical measurement by any eyespot or other different colors. Tests were carried out under same condition: trial wavelength range within 230~800 nm with 0 °incident angle. Reference light incidented with an angle of 8 °. The temperature of the laboratory was at 15~30℃. Relative humidity was less than 65%. Finally, the absorption curves were figured out and the scales with best absorption effect were selected to be examined further.

1.2 SEM observation

Specimens for SEM were taken from the butterfly *Heliophorus ila Lvcaenidae*, which showed the lowest reflectivity. The first step was ungreasing processing. Specimens were rinsed in normal saline solution (0.65% NaCl) and ether for 10 min respectively to get rid of mucus, fat and protein. The second step was dehydration. The ungreased specimens were immersed into series ethanol solution with concentration of 40%, 50%, 70% and pure ethanol at last, 10 min in each solution before natural dehydration. The third step was to paste samples on a metallic substrate with dou-ble-sided conductive tape, make sure not to damage the specimens surface during the whole process. The last step was sput-tering. Finally, all specimens were sputtered with 15~20 nm gold powder within 40 ° rotating angle and examined under field emission scanning electron microscope (FESEM; FEI SIRION 200).

1.3 TEM observation

TEM photos were taken with JEOL-100 microscope system. The specimens were also taken from wings of the butterfly *Heliophorus ila Lvcaenidae* dorsal. The specimens were prepared by the following steps: (1) Immersed in 4% glutaraldehyde for 2 hours to avoid structure change since the evaporation of water; (2) Placed in sodium cacodylate buffer solution for 1.5 hours; (3) Kept in 1% osmic acid for 1.5 hours and dehydrated with ethanol; (4) Embedded in epoxy resin, and solidified for 4 hours with an oven; (5) sliced into lamellas with thickness of 70 nm.

2. Results and discussion

2.1 Spectrometer results

Reflectivity curves of all samples were shown in Fig. 2. The wavelengths ranged from 230 nm to 850 nm and y-axis was the reflectivity (R%).

Three optical effects may be occured when light reaching on the wing surface, namely, reflection, transmission and absorption. The sum of the three portions of light should be 100% as shown in Eq. 1. When the transmission was at a certain level, the lower reflectivity means the higher absorptivity. The absorption capacity could be evaluated by calculating the absorptivity based on the formula.

$$\mathrm{Re}\,flectivity(R\%)+Absorptivity(A\%)+Transmissivity(T\%)=100\% \quad (1)$$

For clear comparison, the curves were displayed in Fig. 2a and 2b separately. In full trial spectrum, Papilio maackii shining area and Apatura ilia exhibited higher reflectance, shown as Fig. 2- Ⅰ and Fig. 2- Ⅴ. This meant their absorptivities were lower. Furthermore, these two curves demonstrated steady tendency and had not any peak value within the whole spectrum. Reflectivity of *Papilio machaon*, *Parnassius stubbendorfii*, *Brenthis daphne*, *Troides brookiana* increased with wavelength obviously, shown as Fig. 2-Ⅱ， 2-Ⅲ， 2-Ⅵ and 2-Ⅶ. The reflectivity value reached to the highest point in the red spectrum. Butterfly *Heliophorus ila Lvcaenidae* (Fig. 2-Ⅳ) had the lowest reflectivity among all the samples. The minimal value plunged to 15%. It means the absorptivity of butterfly *Heliophorus ila Lvcaenidae* could be as

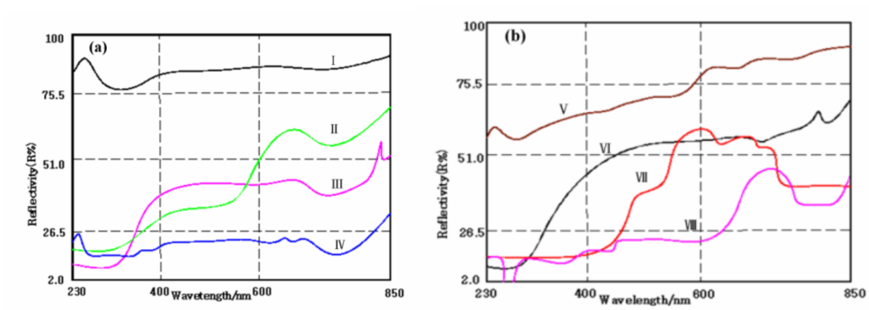

Fig. 2 Spectral curves of several butterfly wings. (a) Ⅰ. butterfly *Papiliomaackii shining area*, Ⅱ. butterfly *Parnassiusstubbendorfii*, Ⅲ. butterfly *Papilio machaon*, Ⅳ. butterfly *Heliophorus ila Lvcaenidae*. (b) Ⅴ. butterfly *Apatura ilia*, Ⅵ. butterfly *Brenthis daphne*, Ⅶ. butterfly *Troides brookiana*, Ⅷ. butterfly *Papilio maackii* black area.

much as 85%. Furthermore, its absorptivity almost remained at a constant level within the entire trial spectrum without any obvious peak.

The reflectivity of the black area of butterfly *Papilio maackii* (Fig. 2-Ⅷ) reached to 50%, which is higher than that of butterfly *Heliophorus ila Lvcaenidae*. the spectral peak emerged at 650 nm. After the primary optical testes and analysis, butterfly *Heliophorus ila Lvcaenidae* scales were selected to be further observed at a micro level with SEM and TEM.

2.2 SEM and TEM results

The overlapping arrangement and morphology of scales could be observed at lower magnification, shown in Fig. 2a. Scales imbricated on the wing surface. There were two types of scales on butterfly *Heliophorus ila Lvcaenidae* wing surfaces. The cover ones (type 1) had regular through-holes structure, through which the incident light passed directly and shined on the ground ones (type 2). The type 2 scales could be observed quite clearly although they hid under cover ones. This confirmed the existence of through-holes of type 1 scales further.

More micro details of a single scale were obtained at higher magnification. Shown in Fig. 3b, there were parallel ridges distributing from fore to end on every scale. There were also many short ribs (the length varied at 100～200 μm) connecting each two adjacent ridges. The distances between each two ribs were almost same, and therefore some rectangular lattices were formed which looked like thousands of window. The regular through-holes structures of the cover scales (Type1 scale) in this manuscript could scatter the incident light, resulting in more and more incident light could irradiate on the surface of the ground scales (Type2 scales). Finally, most of the light energy could be absorbed by the ground scales. The role of the cover scales (Type1 scale) is mainly diffracting light. So, it is largely conducive to absorption instead of direct absorption. There were other kinds of disordered nano-hole structure like this were also found in other butterflies.[29,30]

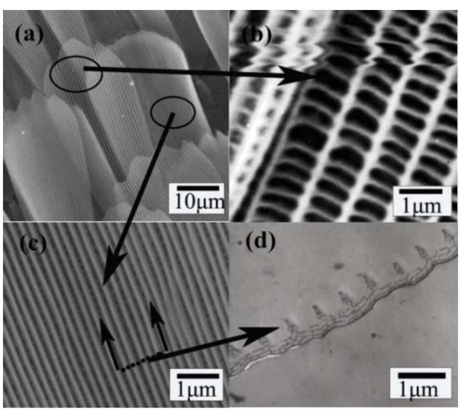

Fig. 3 Images of SEM and TEM. (a) Images of two kinds of scales. The cover ones were type 1 and the ground ones were type 2. (b) Holes in type 1 scales surface. The ridges together with ribs formed holes which the incidence light could pass through. (c) Morphology of type 2 scales, there were ridges occupied more proportion of the lighting area. (d) Cross section image of type 2 scales. Type 2 scales were cut down along the arrows direction in order to obtain the cross section details.

Details of type 2 scales also had ridges (Fig. 3c). However, the difference was that the ridges had rather dense distribution and more greater in width. Spacing between two ridges was about 0.5~0.8 μm. Therefore, ridges occupied more proportion of the lighting area and played a main role in sunlight absorption.

Therefore, even though there were two kinds of scales on butterfly *Heliophorus ila Lvcaenidae* wings, the upper scales had holes structure and rarely absorb sunlight. The underlying scales with ridges have absorption capability.

2.3 Optical Mechanism Analysis

Based on the TEM image of cross section, the optical model of type 2 scales of butterfly *Heliophorus ila Lvcaenidae* were divided into two parts, different optical effects of each part were discussed respectively, (Fig. 3d). The simplified model was built as shown in Fig. 4.

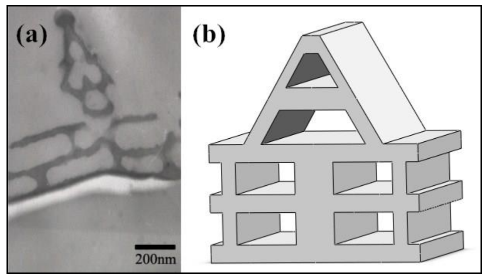

Fig. 4 Microstructure model of scales with absorption function. (a) TEM image of scale microstructure. (b) 3D simplified model.

2.3.1 Multiple reflection and refraction

The upper part was shown in Fig. 5. When sunlight travelled from air (refractive index n_1) to scale chitin material (refractive index n_2) which constitutes original butterfly wing,[31,32] reflection and refraction would happen on the interface at the same time. Some conclusions could be obtained according to the geometrical optics principle: (1) the refraction light exists in the plane of incident light and interface normal; (2) refraction and incident light are separated at each side of the normal; (3) the incidence angle α and refraction angle γ accords with the law as Eq. 2.

$$n_1 * \sin\alpha = n_2 * \sin\gamma \qquad (2)$$

In this case, $n_2 > n_1$. So, $\gamma < \alpha$. The refraction light travelled across the materials and entered into the hollow zone. Then, refraction and reflection occurred at any interface. After multiple reflections and refractions, light travelled for a longer distance, only a small part of light was reflected back to air, resulting in that most of the incident light was effective adsorbed within the structure eventually.

2.3.2 Optical resonator effect of bottom part

Resonant cavity was made up of two pieces of axis vertical plane. The light was trapped into this cavity by reflecting back and forth (Fig. 6). In an ideal resonant cavity, any electromagnetic disturbance behavior would not stop. Electro-magnetic field was calculated by solving Maxwell's equations according to the boundary conditions. The electromag-netic field was considered as a wave bouncing back and forth between cavity walls and resulting in a standing wave field.

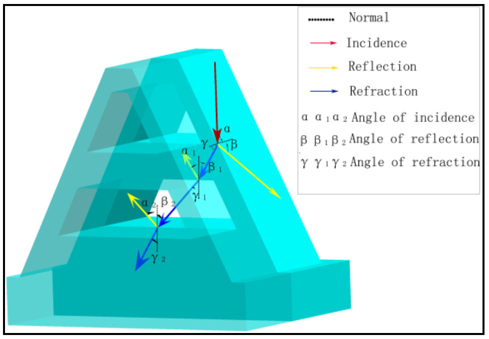

Fig. 5 Schematic of the multiple reflection and refraction occurred on the upper part of the structures of the scale surface.

In this case, the bottom part of scales could be optimized as rectangular cavity of six planes as shown in Fig. 6. The coordinates of the inner surface were shown as Eq. 3.

$$\begin{cases} x=0 \\ x=L_1 \end{cases} \begin{cases} y=0 \\ y=L_2 \end{cases} \begin{cases} z=0 \\ z=L_3 \end{cases} \tag{3}$$

Electric field of electromagnetic wave inner the cavity was E and magnetic field was H, Any rectangular components of E and H should meet the Helmholtz equation. For any component of u (x, y, z), there was $\nabla^2 u + k^2 u = 0$,[33] basing on separation of variables method.Supposed $u(x,y,z) = X(x)Y(y)Z(z)$, then

$$\begin{cases} \dfrac{d^2X}{dx^2} + k_x^2 X = 0 \\ \dfrac{d^2Y}{dy^2} + k_y^2 Y = 0 \\ \dfrac{d^2Z}{dz^2} + k_z^2 Z = 0 \end{cases} \tag{4}$$

$$k_x^2 + k_y^2 + k_z^2 = \omega^2 \mu\varepsilon \tag{5}$$

So, the result of u (x, y, z) was

$$u(x,y,z,) = (C_1 \cos k_x x + D_1 \sin k_x x)(C_2 \cos k_{\partial y} y + D_2 \sin k_y y)(C_3 \cos k_z z + D_3 \sin k_z z) \tag{6}$$

Established boundary conditions were $\vec{n} \times \vec{E} = 0, \partial E_n / \partial_n = 0$ and x=0, y=0, z=0. The arbitrary constant C_i and D_i can be defined, and as a result that $u(x,y,z)$ could be specified as component of $\vec{E}$.

$$\begin{cases} E_x = A_1 \cos k_x x \sin k_y y \sin k_z z \\ E_y = A_2 \cos k_x x \sin k_y y \sin k_z z \\ E_z = A_3 \cos k_x x \sin k_y y \sin k_z z \end{cases} \tag{7}$$

In terms of boundary conditions on planes of $x=L_1$, $y=L_2$, $z=L_3$, the true was that k_xL_1, k_yL_2, k_zL_3 must be integer times of π, namely

$$k_x = \frac{m\pi}{L_1}, k_y = \frac{n\pi}{L_2}, k_z = \frac{p\pi}{L_3} (m,n,p = 0,1,2,\cdots) \tag{8}$$

In Eq. 8, m, n, p represented numbers of half-wave contained in each rectangle side.

After substituting Eq. 7 into $\nabla \vec{E} = 0$, the three arbitrary constants A_1, A_2, A_3 must accord to the relation of Eq. 9.

$$k_x A_1 + k_y A_2 + k_z A_3 = 0 \tag{9}$$

It can be concluded from Eq. 9 that every two constants of A_1, A_2, A_3 were independent. After satisfying Eq. 8 and 9, Eq. 7 could be considered as one sort of region oscillation of electromagnetic field inner the rectangle hollow structure.

For each group of m, n, p, there had two independent polarized wave modes, and the resonant frequency ω can be obtained from Eq. 5 and 10.

$$\omega_{mnp} = \frac{\pi}{\sqrt{\mu\varepsilon}} \sqrt{(\frac{m}{L_1})^2 + (\frac{n}{L_2})^2 + (\frac{p}{L_3})^2} \tag{10}$$

ω_{mnp} was resonant frequency of resonant cavity. The resonance oscillation mode of lowest frequency was wave (1, 1, 0) with wavelength λ shown by Eq. 11.

$$\lambda_{110} = \frac{2}{\sqrt{\frac{1}{L_1^2} + \frac{1}{L_2^2}}} \tag{11}$$

Above was the ideal mode of resonant cavity. In this study, by the butterfly *Heliophorus ila Lvcaenidae*, the scale material consisted of a rectangular cavity. L_1=580 nm, L_2=130 nm, L_3>>L_1, L_2, the materials had a certain energy consumption. The light wave bounced back and forth on the cavity wall. In this process, some portion of light was consumed and absorbed by the materials. Certain light with wavelength λ_{110} was strengthened and reflected back to the air from certain flaws position and produced the wings structural color of butterfly *Heliophorus ila Lvcaenidae*. In this case, the calculation result was λ_{110}=253.7 nm, that's why there was an unobvious peak nearby 250nm spectrum region of curve-Ⅳ in Fig. 2a.

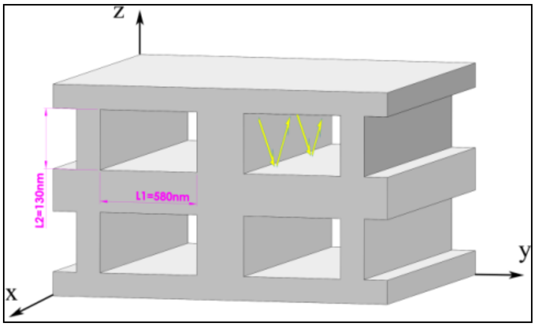

Fig. 6 Schematic of the light absorption occurred on the bottom part of the structures of the scale surface.

3. Conclusions

In this study, the absorption effeciency of seven species of butterflies were tested. It was found that butterfly *Heliophorus ila Lvcaenidae* wings had the highest absorption capability (with absorptivity of 85%) among all samples in spectrum 230 nm～850 nm

SEM results showed that butterfly *Heliophorus ila Lvcaenidae* had two kinds of scales. The sunlight passed through the holes in scales surface and irradiated on the underlying scales. The underlying scales played a main role in the absorbing behavior. TEM analysis illustrated the cross section microstructure of scales type 2. There were two parts of optical delicate nano-architectures. The top part was simplified to a hollow triangle model. Sunlight was reflected and refracted on the slope surface multiply. By this way, the light travelled longer path length in the scale materials. Therefore, more energy was consumed. The bottom part was simplified to a rectangle resonator model. Incidence light was reflected back and forth repeatedly. Energy was absorbed gradually and saved during this endless reflecting process. Only light with wavelength λ_{110}=250 nm could be reflected out to show the structural color of butterfly *Heliophorus ila Lvcaenidae*.

This study focuses on the absorption mechanisms inner microstructures of the scales. Contents of this paper has an important reference value for solar heat utilization research via bionic way.

Knowledgment

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