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Mouchet, Sébastien; Lobet, Michaël; Kolaric, Branko; Kaczmarek, Anna; Van Deun, Rik; Vukusic, Peter; Deparis, Olivier; Van Hooijdonk, Eloïse

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Photonic scales of *Hoplia coerulea* beetle: any colour you like[★]

Mouchet S. R.^{a,b,*}, Lobet M.^b, Kolaric B.^{b,c}, Kaczmarek A. M.^d, Van Deun R.^d,
Vukusic P.^a, Deparis O.^b, Van Hooijdonk E.^b

^aSchool of Physics, University of Exeter, Stocker Road, Exeter EX4 4QL, United Kingdom

^bDepartment of Physics, University of Namur (UNamur), Rue de Bruxelles 61, B-5000 Namur, Belgium

^cCurrently with Micro- and Nanophotonic Materials Group, Faculty of Science, University of Mons, Place du Parc 20, B-7000 Mons, Belgium

^dL³ – Luminescent Lanthanide Lab, Department of Inorganic & Physical Chemistry, Ghent University, Krijgslaan 281-S3, B-9000 Ghent, Belgium

Abstract

The scales covering the elytra and thorax of the male *Hoplia coerulea* scarab beetle are a specific case of natural photonic structure encased by a fluid-permeable envelope. These scales display several optical effects including structural colouration, fluid-induced colour changes, fluorescence emission and fluid-induced fluorescence changes. Although the photonic structure is not directly exposed to the environment, optical changes are known to be induced by liquids and vapours. These changes are controlled by the envelope that mediates exchanges with the surrounding environment. The optical system comprising the photonic structure and the envelope was previously termed “photonic cell”. Novel possibilities to develop functional photonic materials and smart coatings are offered by such a system through a bioinspired approach. This technological potential motivated us to analyse theoretically, through numerical simulations, the water-induced changes of optical properties of light emitters embedded in a photonic structure inspired by *H. coerulea* beetle’s scales.

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Keywords: Structural colours; photonic crystals; photonic bandgap materials; beetle scale; natural photonic crystal; fluorescence; colour; optical sensors;

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* Corresponding author. Tel.: +44 (0)1392 724156.

E-mail address: s.mouchet@exeter.ac.uk

1. Introduction

In nature, several cases of photonic structures encased by a permeable envelope can be found [1-3]. This is the case of the structure at the origin of the blue-violet colour of the scales covering the elytra and thorax of the male *Hoplia coerulea* scarab beetle, also known as the cerulean chafer beetle. This insect from the family Scarabaeidae is found in Northern Spain and Southern France. Although the male beetle displays a blue colouration (Fig. 1), the female is dull brown due to pigments (Fig. 2a-b). Ventral scales and the ones covering the legs of the male and the female are greenish white. This insect has been known since the 18th century when its shiny blue appearance was described by entomologists including Drury and Voet [4,5]. Historically, the beetle was called *Scarabaeus coerulea* meaning “blue beetle” (Drury, 1773) or *Venator coeruleus*, “blue hunter” (Voet, 1776) [4,5].

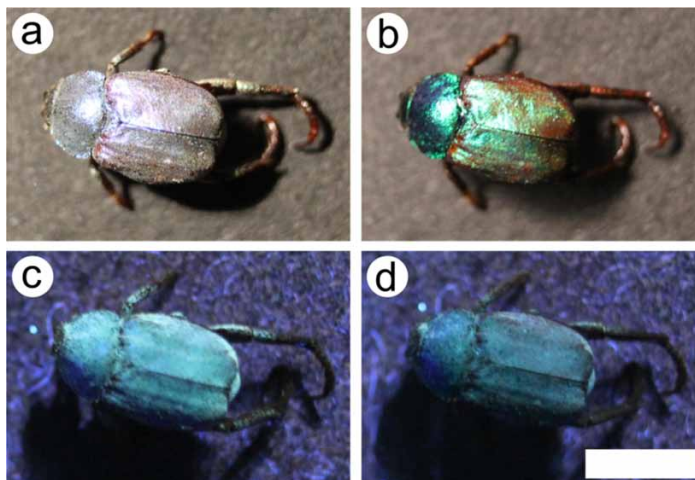


Fig. 1. Water-induced colour and fluorescence changes of the elytra and thorax of the male *Hoplia coerulea* beetle. Under visible white light, the beetle elytra appear blue-violet in the dry state (a) and green in the wet state (b). Illuminated by ultraviolet light, a turquoise colour is displayed in the dry state (c) and a navy blue colour in the wet state (d). Scale bar: 5 mm.

Following these early observations, the scales of this beetle were shown to exhibit a diversity of optical properties: structural colours [6-8], colour changes induced by contact with liquids [9-12] and vapours [10,13], fluorescence controlled by the photonic structure [14] as well as changes in fluorescence emission induced by liquids [12].

The beetle scales have an approximated diameter of 80 μm and a thickness of about 3.5 μm . The origin of the optical effects is a periodic porous multilayer encased by a 100 nm-thick envelope [8]. The photonic structure, which controls both the insect's colouration and fluorescence, is therefore not open to the surrounding environment. Because of the role of this envelope, the photonic system comprising the multilayer and the envelope was called a “photonic cell” [12,13], i.e., a biocompatible photonic structure enclosed by a permeable envelope mediating fluid-induced optical changes in the structure.

The liquid-induced fluorescence emission change in this porous photonic structure offers a new possibility to develop bioinspired functional photonic materials and smart optical coatings based on the mechanism highlighted by these investigations. This luminescence in natural photonic structures will certainly inspired applications in various fields such as sensing and solar cells as well as in medical therapy and diagnosis [15].

Here, we review, in this article, the literature on the optical properties of this insect before investigating the optical properties of a bioinspired analogous model.

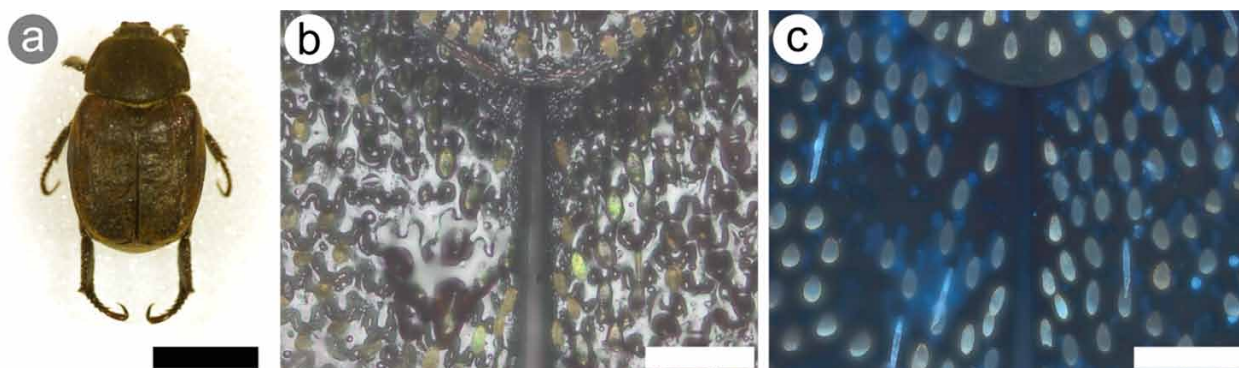


Fig. 2. (a) The female *H. coerulea* beetle displays a dull brown colour. Its elytra are covered by very small orangish brown scales (b) that give rise to fluorescence emission under UV light (c). Scale bars: (a) 5 mm; (b-c) 200 μm .

2. Photonic structure in the scales of male *H. coerulea* beetle

The vivid blue-violet colour of the male *H. coerulea* beetle is metallic (i.e., incident light is specularly reflected by its elytra) and iridescent (Fig. 3) [6-8]. The Bragg reflectance peak position blue-shifts from about 460 nm (i.e., blue colour) to 400 nm (i.e., violet colour) when the incidence and detection angles (measured with respect to the normal to the sample) increase from 0° to 60° (Fig. 3b). Since the study by Vigneron *et al.* [8], this colour is known to arise due to a photonic structure located within the scales covering the insect's body. This structure comprises a macroporous periodic multilayer composed by 35 nm-thick dense planar layers and 140 nm-thick macroporous spacer layers encased by a 100 nm-thick envelope (Fig. 4). Due to the specular reflection by the elytra, the photonic structure was modelled as a 1D photonic crystal comprising 12 chitin layers ($n_{\text{chitin}} = 1.56$) and 12 spacer layers, the refractive index of which was calculated by an effective medium approximation ($n_{\text{mixed}} = 1.26$). Iridescent coatings inspired by this photonic structure were designed. They comprised a periodic stack of TiO_2 and SiO_2 layers deposited on glass substrates by magnetron sputtering [16].

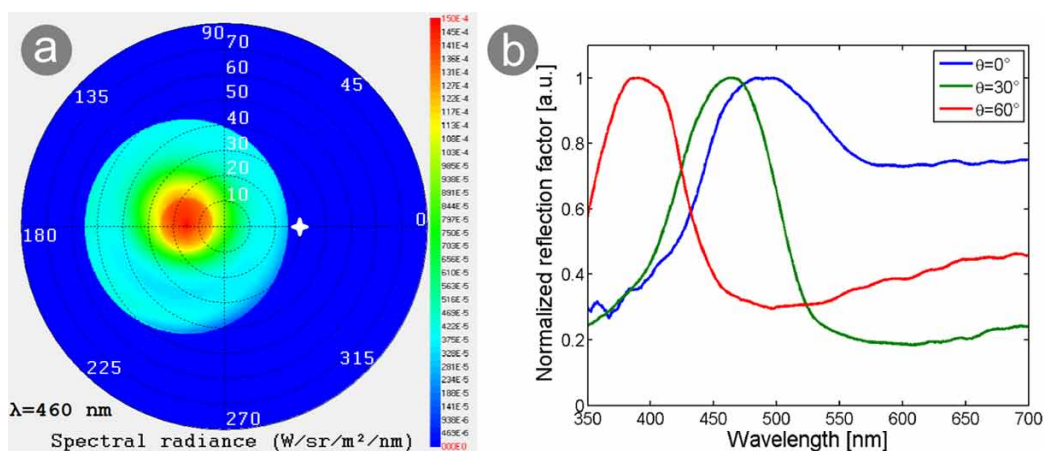


Fig. 3. (a) Light reflection on *H. coerulea* elytra is specular and very directional as shown by this spectral radiance map, measured within a solid angle with polar and azimuthal angles ranging from 0° to 80° and from 0° to 360° , respectively. The incident light beam (460 nm-wavelength) formed a 30° -angle with the normal to the elytra (the white star represents the incident direction). (b) *H. coerulea*'s colour is iridescent: the reflectance peak wavelength blue-shifts with an increase of the incidence and detection angles.

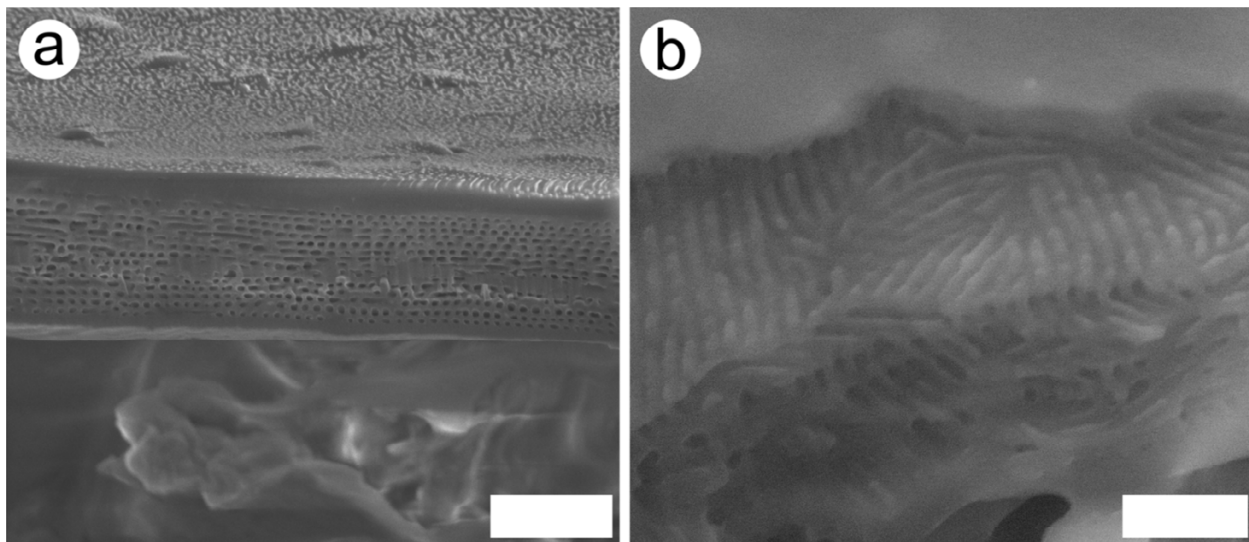


Fig. 4. (a) The porous multilayer structure found in the scales of male *H. coerulea* beetle is at the origin of the blue-violet iridescent colour of the insect. The structure comprises a periodic stack of chitin layers and spacer layers encased by a 100 nm-thick envelope. (b) The spacers are not sufficiently organised in order to give rise to a diffraction grating. In the optical model, the spacer layers were assumed to be homogeneous layers with an effective refractive index. Scale bars: (a) 2 μm ; (b) 1 μm .

3. Fluid-induced colour changes

A few years ago, it was highlighted that the beetle's colour reversibly changes from blue-violet to green when the elytra are immersed in water (Fig. 1a-b) [9]. These changes are explained by the filling of the structure macropores with water ($n_{\text{water}} = 1.33$), leading to the increase of the effective refractive index of the spacer layer ($n_{\text{mixed}} = 1.45$ instead of 1.26) and to the red-shift of the reflectance peak position as well as to the decrease of the reflectance peak intensity [9]. More recently, it was demonstrated that if droplets of different liquids (distilled water, methanol, absolute ethanol, 2-propanol, propanone, acetonitrile, methylbenzene or ethoxyethane) are deposited on one elytron, the colour changes are somewhat similar: one by one, the scales turn from blue-violet to green [12]. In the case of methylbenzene, which possesses a significantly higher refractive index than the other liquids tested (approximately 1.50 as opposed to 1.33-1.38), the green colouration is darker, because of the more striking increase of the effective refractive index of the spacer layer. The main difference that has been observed between these liquids is the time taken for the changes [12]. Despite the envelope preventing direct exposure of the structure to fluids, the colour of the beetle changes upon contact with liquids. These changes were explained by liquid penetration through the encasing envelope towards the photonic structure, the structure and its microporous envelope being made of the same chitin-based material [9,12].

As in other natural photonic structures [17-25], the reflectance changes of the beetle's cuticle were also observed using vapours [13]. The beetle's scales were shown to be more sensitive to water vapour than to ethanol vapour [13]: a significant red-shift of the reflectance peak was observed when the elytra were in contact with a water vapour concentration higher than 50% and with an ethanol vapour concentration higher than 75%. Another feature that was noticed is the increase in intensity of the reflectance peak. This feature is not commonly associated with fluid-induced colour changes and is explained by the filling of the micropores within the body of the photonic structure, giving rise to an increase of the refractive index contrast between the spacer layers and the chitin layers [13].

However, these fluid-induced colour changes turn out to be perplexing. On one hand, the colour changes appear to be less sensitive to ethanol than to water. On the other hand, the affinity of the cuticular material composing the beetle's scales, quantified in terms of contact angle, was shown to be higher with ethanol (0° contact angle) than with water (about 76°) [12]. This behavioural difference may seem counterintuitive: in spite of the fact that the contact angle of water droplets is higher than for ethanol droplets, the penetration of water is faster. These puzzling

observations are explained by taking into account physico-chemical properties of the cuticle material. Indeed, it was found that chitin is more permeable to water than to ethanol [26,27]. Furthermore, the presence of salts (NaCl and KCl) was found in the scale material by X-Ray Photoelectron Spectrometry (XPS) analyses [12]. These salts also influence the permeability of the material.

The envelope of the scales forms a barrier mediating fluid exchanges with the surrounding environment and subsequently controls the colour changes. Given the resemblance with the role of a plasma membrane which mediates the exchanges of a biological cell with the environment, the natural system comprising the photonic structure and the encasing envelope was termed a “photonic cell”. This concept is interesting for the development of applications such as vapour sensors, smart glass windows or monitoring cell-metabolism in culture media [28–32].

4. 4. Fluorescence and fluid-induced fluorescence changes

Like many animals such as arthropods, birds or marine organisms [33–36], *H. coerulea* beetle gives rise to fluorescence emission when it is illuminated by ultraviolet light [12,14] (Fig. 1c and 2c). This emission of light originates from the presence of fluorophores naturally embedded within the living organism's teguments. The confinement of fluorescent emitters within photonic structures enables the so-called controlled fluorescence, which is known to modify properties such as the intensity (enhancement/inhibition) or spatial distribution (directivity of emission) of the fluorescent response [37–40]. Such a photonic confinement effect is found in several living organisms [41,42]. In the case of the investigated beetle, the fluorescence emissions of the male and female scales are turquoise (Fig. 1c) and light blue (Fig. 2c), respectively.

Following contact with water, changes of fluorescence emission from turquoise to navy blue (Fig. 1c-d) were observed with the male beetle [12]. The changes were also explained by the filling of the structure macropores. These observations may be of great interest for the design of novel optical systems in lighting, imaging, biosensing and theranostic nanomedicine (e.g., safety signs based on long persistent phosphors and photonic structures or fluorescent trace detectors combined with photonic structures) [15,43–45].

Hereafter we investigated the optical emission from a photonic system model inspired by the *H. coerulea*'s scales in addition to the changes induced by water. This model comprises the photonic structure, as described by Vigneron *et al.* [8] and Rassart *et al.* [9], in which fluorophores (or any other kinds of light nanoprobe such as metallic or semiconductor nanoparticles and quantum dots) were assumed to be embedded in the bottom layer of the structure. Therefore, a planar emitting source was located in this bottom layer. The bioinspired analogous model will be easy to synthesise with standard deposition techniques (e.g., thermal deposition, physical vapour deposition or atomic layer deposition).

The simulations enabled us to visualize the propagation of internally emitted light within the photonic structure. A remarkable difference in the electric field propagation (real part of the z -axis component) is predicted between the dry and the wet states (Fig. 5a-b). When light is emitted at a wavelength located in the bandgap of the dry state (Fig. 5a), it can barely couple out of the structure. On the other hand, when the photonic structure is in the wet state, propagation is allowed at $\lambda = 459$ nm (Fig. 5b) because this wavelength is outside the bandgap of the photonic structure in the wet state [12]. Actually, this wavelength is close to the blue edge of the photonic bandgap (474 nm), where light benefits from enhanced transmission due to slow light effect [46,47].

When the source wavelength is located at the blue edge of the bandgap, the electromagnetic energy concentrates in the thick effective spacer layers (of lower refractive index) whereas at the red edge, a substantial part of the energy concentrates in the thin dense planar layers (of higher refractive index) [48]. Indeed, since the distribution of the electromagnetic energy density is determined by the refractive index contrast ($n_{\text{chitin}}/n_{\text{mixed}}$), when this contrast is large, the energy is preferentially localised in either the dense planar or the effective spacer layers according to the side of the bandgap where the source wavelength is located. However, when the contrast is small, the energy density distribution is more uniform across the layers, which is the case when the structure is impregnated by water. Following the decrease in refractive index contrast induced by water, the energy density distribution shifts from one layer type to the other one (Fig. 5c). At a wavelength of 459 nm, the energy density in the chitin layers is consequently lower in the wet state than in the dry state (Fig. 5c). Quantitatively, the ratio $r = U_{\text{chitin}}/U_{\text{mixed}}$ of the energy densities in the dense planar layers and in the effective spacer layers reduces from $r = 0.31$ to 0.26.

This numerical study presents a first step towards the fabrication of bioinspired analogous photonic cells embedding fluorophores and turns out to be interesting for the design of applications through a bioinspired approach.

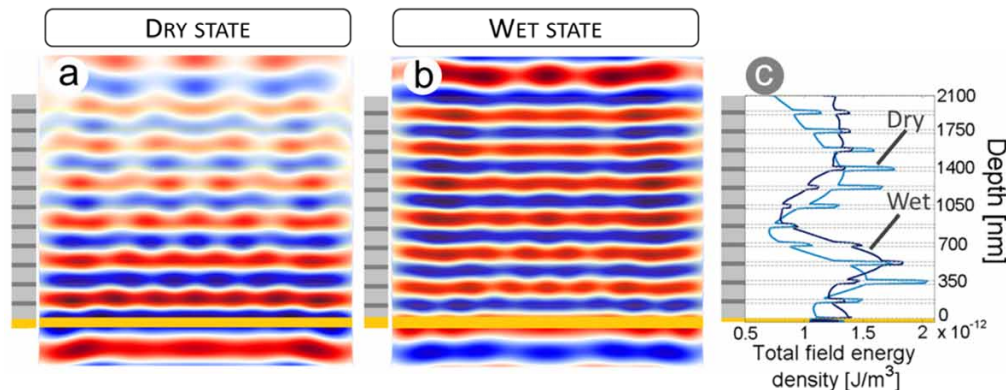


Fig. 5. Spatial distribution of the z -component of the electric field with emission at $\lambda = 459$ nm in the dry (a) and in wet states (b) of the photonic structure. A schematic representation of the photonic structural model with the spectrally uniform planar source located in the bottom layer (yellow), the dense planar layers (dark grey) and the effective spacer layers (light grey) is inserted. (c) Profiles of the total field energy density along the depth of the structure in dry and wet states.

5. Conclusion

The photonic structure of male *Hoplia coerulea* beetle is surprising since it gathers several optical properties in a single entity: structural colours, fluid-induced colour change, fluorescence emission and change of fluorescence emission enacted by liquids. The photonic structure consists of a 1D periodic porous multilayer encased by an envelope. Although the structure is not open to the surrounding environment, optical changes induced by liquids and vapours are observed. These changes are controlled by the permeable encasing envelope that mediates exchanges with the environment. This system comprising the photonic structure and the envelope was previously termed “photonic cell”. We investigated numerically the emission properties of light emitters embedded in such a photonic cell inspired by *H. coerulea*’s scales as well as their changes induced by water. Due to the shift of the photonic bandgap upon contact with water, both the emission of light by a source embedded in the structure, and the distribution of the electromagnetic energy density of the propagating fields are modified. Such an optical device offers a new possibility to develop novel functional photonic materials and coatings in areas such as sensing and solar cells as well as medical therapy and diagnosis.

Note on methods

Pictures of *H. coerulea* beetles were taken with a EOS 1100 D Canon camera equipped with one EF92 II Canon extension tube under an IKEA Typ A9904 white halogen lamp (for the visible light observations) and an UV Midlight Labino lamp (for the fluorescence observations) in both dry and wet states. The wet state was reached by immersion of the beetle into water for 15 min. The background was a black sheet of paper (for the male beetle) and white foam (for the female).

Optical microscopy was performed thanks to an Olympus BX61 microscope equipped with an Olympus XC50 camera with an Olympus BX-UCB white light source and a Lumen Dynamics X-cite Series 120PCQ UV lamp.

The spectral radiance map was measured with an ELDIM EZ Contrast XL80MS scatterometer with an incidence wavelength equal to 460 nm and an ELDIM EZ Reflex Source 300 W light source. The measurements were performed within a solid angle with polar and azimuthal angles ranging from 0° to 80° and from 0° to 360°, respectively. The incident light beam formed a 30°-angle with the normal to the elytron surface.

The normalized reflection factor was measured with an Avantes AvaSpec-2048-2 spectrophotometer and an AvaLight-DH-S-BAL deuterium-halogen light source. The reflected intensity was measured with respect to an

Avantes WS-2 white diffusor. The different incidence and detection angles were fixed by an Avantes AFH-15 fiber holder.

The electron microscopy observations were performed with a Fei Nova Nanolab 200 Dual-Beam scanning electron microscope (SEM) and a Fei Quanta 200F SEM.

The propagation of the electromagnetic field was computed with the finite-difference time-domain (FDTD) method [49] using the MIT Electromagnetic Equation Propagation (MEEP) package [50]. These simulations were performed with perfectly matched layers (PML) as boundaries as well as a Gaussian source emitting continuously and characterised by a frequency width equal to 0.1% of the central frequency (corresponding to a wavelength equal to $\lambda = 459$ nm).

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