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## Precious Coral

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### Abstract

Precious coral, with attractive pink-to-red color, has been used for ornamental purposes for several thousand years. According to the related science, attractive red precious corals material, are defined to the class Anthozoa, subclass Octocorallia, order Alcyonacea, suborder Scleraxonia, and family Coralliidae in zootaxy. About 30 species are discovered in Coralliidae family compared to the huge Cnidaria phylum. *Corallium rubrum*, *Corallium japonicum*, and *Corallium elatius* are the three main species in Coralliidae family used for jewelry material in the gem market. The purpose of this chapter is to show the nature of animals in Coralliidae family, analyze the nondestructive test methods to identify the natural species from the imitations, and discuss the origin of color and the interactions in between the organic matrix and mineral. The chapter was organized in six parts. The first part reviews the history of precious coral used for different purposes by humans and then describes exact affiliation of precious coral on zoology and taxonomy. The second part deals with the biology and formation of precious coral. The paragraph also presents the information about Coralliidae colonies' sexual maturity, life span, growth rate, and mortality. The trade market and conservation are also summarized in this part. The gemological properties of *C. rubrum*, *C. japonicum*, and *C. elatius*, the main species in precious coral market, are introduced in the third part. As a consequence, an effective and nondestructive identification method to distinguish natural precious corals from their imitations was stated with Raman spectra as demonstrated in the fourth part. In the fifth part, the origin of precious coral color based on the results of Raman scattering measurements and PeakFit analysis is demonstrated. Three different excitation wavelengths (785, 633, and 514 nm) were used for the same samples at the same points. The result shows that all of the samples are colored by a mixture of pigments. Different colors are explained by different mixtures, not by a single pigment. Organic composition, even present in a small amount, plays an important role in the color of precious corals. The sixth part concludes the text by presenting what we have

learned from the experimental data of microscope, SEM, TEM, and EBSD. The spatial relationship between the organic matrix and mineral components is determined by SEM observation on decalcification-treated samples. By integration of the results from nanometer to centimeter-scale detections, a hierarchical structure of precious coral is revealed. In the skeleton of precious coral, building blocks are arranged into several hierarchical levels of oriented modules. The modules in each hierarchical level assemble into larger unit that comprises the next higher level of the hierarchy. Precious coral, as a member of biomineral family, assembled skeleton as a delicate arrangement of a hierarchy of crystals with well-defined orientations under the control of organic matrix. Organic matrix works in both color pigment and architecture field for the precious coral.

**Keywords:** precious coral, organic matrix, oriented module

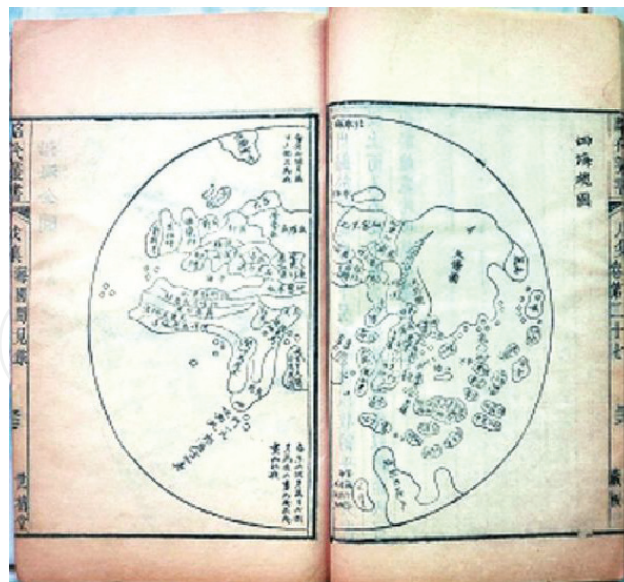
## 1. Introduction

Precious coral, as an organic gem material with attractive pink-to-red color, has been used for ornamental purposes for several thousand years. The earliest history of this material used for decoration could be traced back to 8000 BC for the red coral amulets uncovered in Neolithic graves in Switzerland. It was also reported the discovery of fine coral jewelry in Sumeria and Egypt around 3000 BC [1]. Coincidentally, precious coral was valued highly in Asian Culture throughout history.

In China, precious coral was recognized treasure of the ocean since Three Kingdoms Times around 2000 years ago. It appeared constantly in palace tribute, imperial trappings, Buddha beads, medicinal material during the complete China's history after then. The first description about precious coral was the geographic monograph of the South China Sea with the book name "*Fu Nan Zhuan*". The morphological characteristics of coral were demonstrated in that ancient writing (**Figure 1**). Later, precious coral was also mentioned as medicinal material in literatures of both Li Jing (Tang Dynasty, 659 AD) and Li Shizhen (Ming Dynasty, 1578 AD). Due to its distinctive tree-like appearance and hard texture, precious coral was mistaken for plant or mineral for its early history. The misunderstanding about the nature of precious coral was not by chance, even distinguished naturalist Pliny the Elder named it *Zoophyte*, which habituated with the features of both animal and plant.

Being benefited from the developments of biology and taxonomy, the biological behavior and habits on coral's growth, reproduction, nutrition, and habitat are much well known nowadays than before. According to the related science, attractive red precious corals, which are commonly used as jewelry material, are defined to the class Anthozoa, subclass Octocorallia, order Alcyonacea, suborder Scleraxonia, and family Coralliidae in zootaxy [2].

Since fine specimens of red coral, which is also called gem-class coral or precious coral (**Figure 2**), are the most desirable yet among the least available [3], series of enhancement and treatment technologies, such as dyeing, bleaching, and polymer impregnation, are applied to precious coral's imitations to get higher value. Since a significant amount of imitations share the similar color distribution and structural characteristics with the natural precious corals, the



**Figure 1.** “Fu Nan Zhuan” in Three Kingdoms Times described the morphological characteristics of coral for the first time.

traditional identification methods are facing the difficulties to distinguish the precious corals from the others. The present chapter looks the current statues of precious coral from zoological taxonomy, gemological properties, and adopted methods to explore the essence of precious coral’s attractive color and to determine the spatial relations between the organic matter and mineral component in order to provide information for identification, appraisal, and biomineral research.



**Figure 2.** Fine precious coral carving designed as religious blessing pattern.

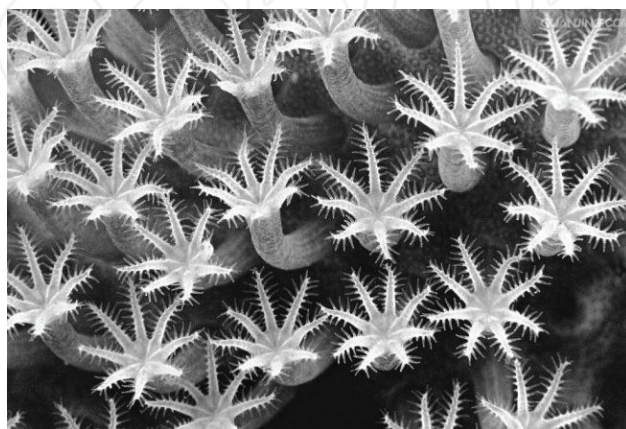
## 2. Biology and formation

### 2.1. General biological background of precious coral

The term coral, as it is commonly intended by divers and enthusiasts, leads itself to numerous misunderstandings in strictly zoological terms. This happens mainly because many books on these extraordinary organisms broadly use the word coral to describe all creatures with a hard skeleton. This definition may thus be used to group organism very different one from the others. The term precious coral ought to be used only for the Coralliidae family in zootaxy. About 30 species are discovered in Coralliidae family compared to the huge Cnidaria phylum, which is containing over 10,000 species of animals.

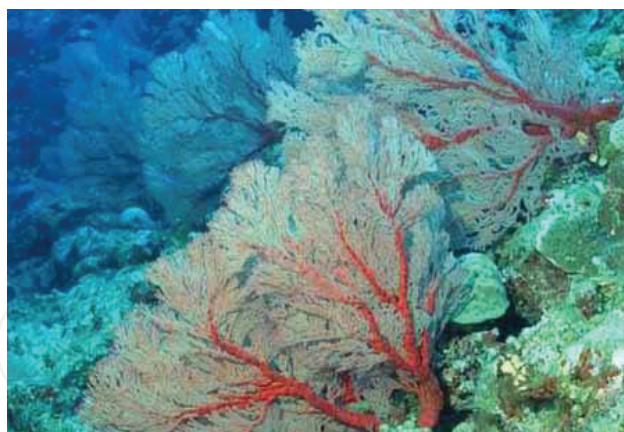
Despite the obvious differences in form, shape, and dimension between one species and another, animals in Coralliidae family share the same basic structural distinction. They keep the sedentary living habits of organisms, mobile only at the larva stage. Every animal can be divided according to the infinite patterns of symmetry along the radius of incredible regular circle. The animal responsible for skeletal formation is precious coral's primary unit-polyp, which is a sort of sac adhering at the base to a rigid substratum and having an aperture facing upwards, surrounded by a variable number of tentacles (**Figure 3**). The number of these (six, eight, or multiples thereof) allows the Anthozoa group to be divided into Octocorallia or Hexacorallia. The Coralliidae belongs to Alcyoniina suborder, Alcyonacea order, and Octocorallia subclass accordingly.

Precious corals are colonial animals, which consist of thousands of small polyps. They reproduce sexually by releasing eggs and sperm polyps of gametes of the same species simultaneously over a period of one to several nights around a full moon. The polyps remain connected and continue to grow and reproduce on their own. These colonies can live for several centuries, during which their continuous calcification creates layered skeletal archives that used to be the material for the jewelry, medicine, and so on. The formation process of the precious coral's skeleton inspired the understanding on bio-mineralization and environmental impact on biological growth. Coralliidae corals grow under strict habitat requirements, which include deep water, rocky bottom, typically aggregate on banks, seamounts, under ledges, and



**Figure 3.** The arrangement of the polyps of coral colonies.





**Figure 4.** Coralliidae corals inhabit deep water, rocky bottom habitats.

in and around caves (**Figure 4**); generally, where there are strong bottom currents. The suitable growth temperature for precious corals is from 8 to 20°C.

Coralliidae colonies reach sexual maturity until 10–12 years old after then adhere to the bottom. Compared with other invertebrate animals, Coralliidae colonies live a long life, grow at slow rate, and die at low mortality. S. Giacomelli, F. Cicognà, and G. Bavestrello studied the biology of Coralliidae colonies from 1965 to 1966. The results indicate that the colonies can attain height of only 1 cm during 1 year. It is impossible to artificially breed the Coralliidae colonies until nowadays [4].

## 2.2. Trade market

The traditional trade market of precious coral was concentrated in Italy at the very beginning, which was supposed to be the earliest and biggest market once upon a time. There was significant trade in precious coral between the Mediterranean and India. Italy, as an irreplaceable trade market, stood head above the other trade regions could be boiled down to its unique integrated functions of precious corals' producing region, design center, and commercial market. A creative drum-like precious coral bead was first designed in Italy and became widely spread to Tibet and Japan along the Silk Road. The trade market on precious coral's season began and lasted for several centuries.

In 1847, precious coral was explored in Sea of Japan infused the fresh blood to the market. As a result, the output production of precious coral went straightly upwards while the fine designed jewelry was more popular than ever before. The history occurred similarly in 1923, a new fishery of precious coral was discovered in Taiwan that year. After the development over decades, Taiwan caught up from behind to be the world's biggest producing region for precious coral in 1964. It was reported that the output of precious coral in Taiwan accounted for 80% compared with the total production of the world in 1984. Over 90% precious corals produced in Taiwan were exported to Japan and Italy during that time.

The red coral species produced in Italy grew in relatively shallow region of the sea. The diameter of the Italian precious coral was small with a diameter less than 10 cm as the consequence of its

habitat, which provided easier conditions for collecting. It explained that the common coral jewelry style in Italy was delicate, exquisite, and always set in gold or silver. Precious coral species grew in Taiwan sea were in the depth of over 200 m and could reach the size of several meters. Thus, the jewelry designs of precious coral vary from sculpture, carving, snuff bottle, headwear to common jewelry. Since precious coral is a non-renewable resource, many regulations or bans on protection are launched by governments all over the world.

### 2.3. Conservation

Since the species of Coralliidae family could not be cultivated, the limited resources raised public concerns on their protection. Recently, on April 8, 2008, China, which now has domestic laws to protect these species, requested that CITES include four *Corallium* species (*C. elatius*, *C. japonicum*, *C. konojoi*, and *C. secundum*) under Appendix III [5].

Meanwhile, the rising price of precious coral resulted in a variety of imitations is flooding the market. Consequently, the research on the composition and structural properties of precious coral, which will promote the development of identification of precious corals from their imitations, is more desired for both the researchers and public than ever before.

## 3. Basic gemological properties of main commercial species in Coralliidae family

*C. rubrum*, *C. japonicum*, and *C. elatius* are the three main species in Coralliidae family used for jewelry material in the gem market. Since they are belonging to the same zoological family, they have some components or structural characteristics in common and also some distinctive features of each own (Table 1).

### 3.1. Chemical composition and mineral components

As being determined by EPMA, LA-ICP-MS, FTIR, and XRD, the samples from Coralliidae family show similar principle chemical composition of  $\text{CaCO}_3$  and major mineral component of high-Mg calcite (Figures 5 and 6). Beyond that, the samples also contain a small amount of organic matters, which play a magic role in the construction of precious coral skeleton (will be explained in Sections 4 and 5). One interesting fact is the theory several decades ago indicated that the color of precious coral was caused by metal ions such as Cu and Fe, of which they absorbed from the seawater, is negated by the test results of EPMA with small to non-detective amount of these elements (Table 2).

### 3.2. Optical properties

#### 3.2.1. Color

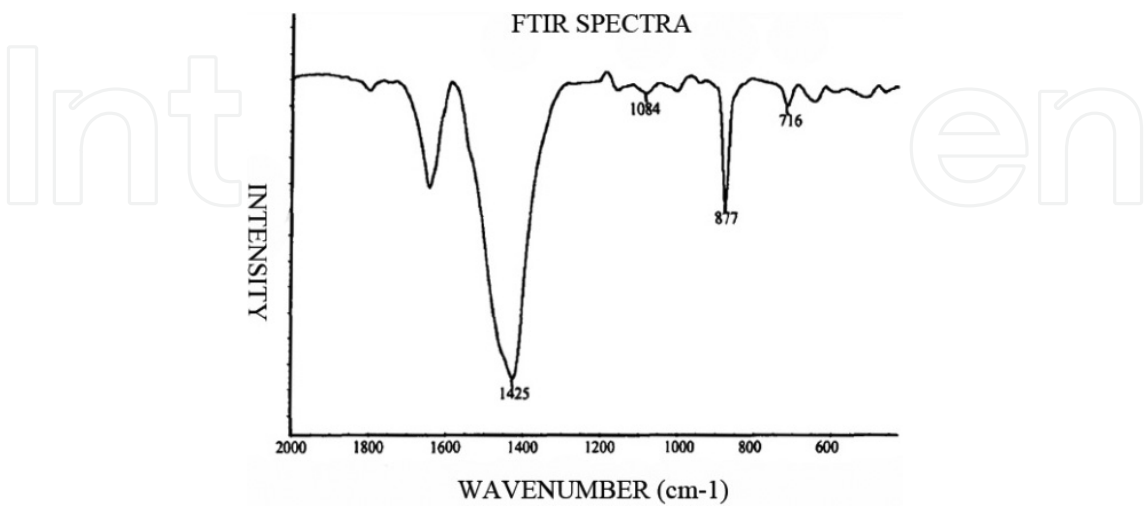
Most precious corals show attractive even pink-to-red color, while some species have white spots, white core, or entire white on the body. To conclude, three main precious coral species'

Trade name	Biological term	Producing area
Aka Coral, Aka Red Coral	<i>C. japonicum</i>	Taiwan, Japan
Momo Coral, Formosa Coral, Pink Coral	<i>C. elatius</i>	Taiwan, Japan
Midway Coral	<i>C. secundum</i>	Midway
Deep Sea Coral	<i>Corallium</i> sp.	Midway
Sardinia Coral, Red Coral	<i>C. rubrum</i>	Southern Europe, North Africa
White Coral	<i>C. konojoi</i>	Pacific Ocean

**Table 1.** Main precious coral species’ trade names and biological term.



**Figure 5.** *C. japonicum* always has glass bright luster.



**Figure 6.** The FTIR spectra of precious samples identifies the biogenic calcium carbonate phase of the skeleton ( $\text{CaCO}_3$ ), with peaks positioned at 1084, 817, and 716  $\text{cm}^{-1}$ .



Composition elements ( $\omega_B\%$ )									
	Na	Ca	Mn	Mg	Ba	Fe	Si	Cu	Sr
Max	0.503	38.775	0.015	4.413	0.161	0.381	7.652	0.0752	1.063
Min	0.227	28.022	—	0.108	—	—	—	—	0.192

**Table 2.** The value of principle composition elements in precious corals.

features of color are described in **Table 3** and **Figures 7–9**. The features of color are the important information to distinguish the different species of precious coral when they are kept of original dendritic shape.

3.2.2. Luster and transparency

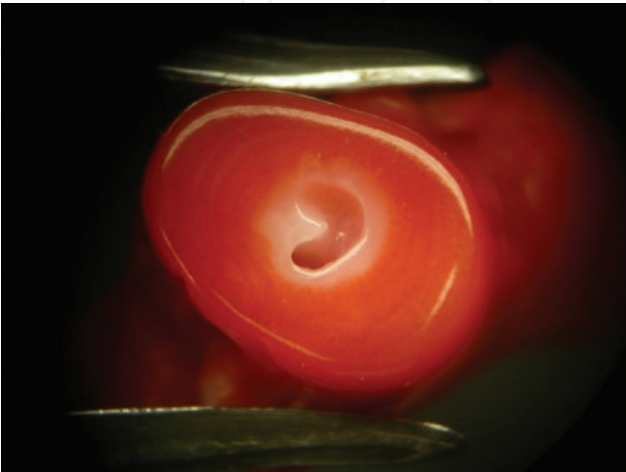
Precious corals show waxy to glass luster when they are polished. Among the three species of precious corals, *C. japonicum* obtained the best transparency of subtranslucent, while *C. elatius* and *C. rubrum* are opaque.

3.2.3. Refractive index

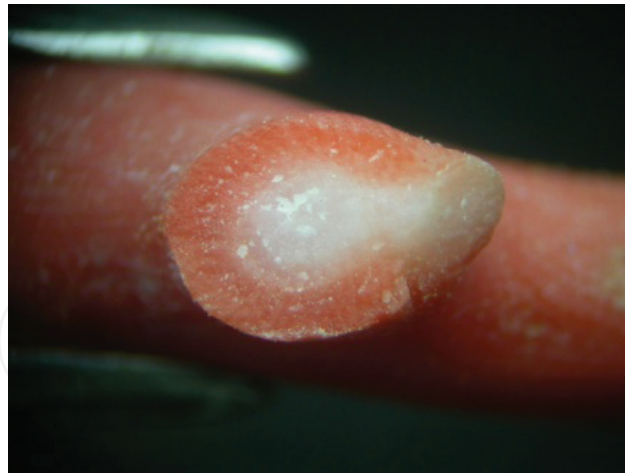
The refractive indexes of precious corals are ranged from 1.49 to 1.65.

Species	Color features
<i>C. japonicum</i>	Orange-to-red color has white core certainly
<i>C. elatius</i>	Pink-to-red color, some specimens are white entirely, white core appears in most specimens
<i>C. rubrum</i>	Deep red color does not have white spot or core

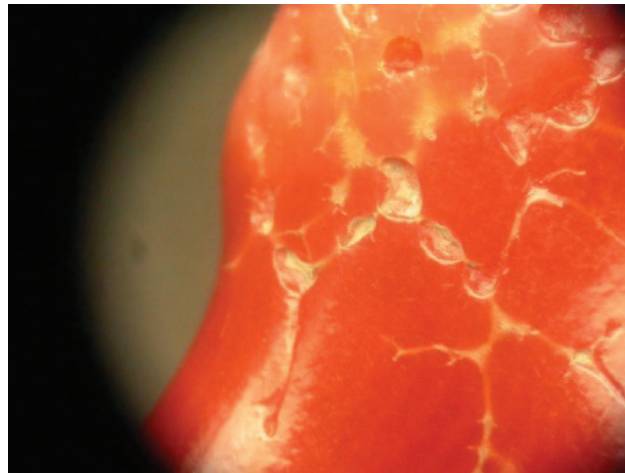
**Table 3.** Precious coral species’ color features.



**Figure 7.** White core of *C. japonicum*.



**Figure 8.** White core of *C. elatius*.



**Figure 9.** Worm cavities distributed on *C. rubrum*.

### 3.3. Mechanical properties

#### 3.3.1. Hardness

Moh's hardness of precious corals is 3–4. However, brittleness of precious coral species is determined by environmental factors like ocean depth they grow at. Some data indicate that the precious corals, which living in deep sea, are easier to get fragile.

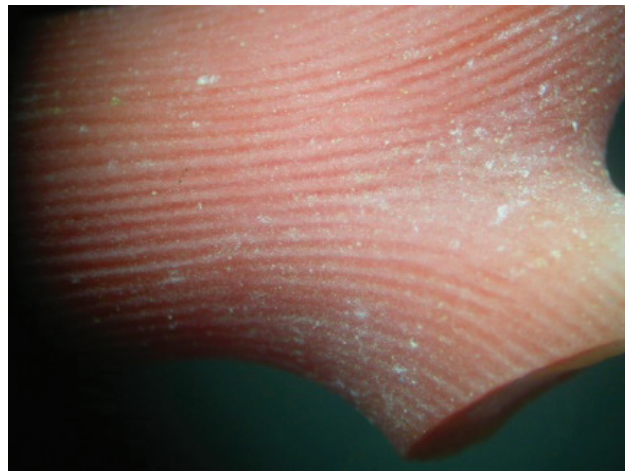
#### 3.3.2. Relative density

As tested by hydrostatic weighing method, the results show the slight difference among the three species of precious coral. The relative density of *C. japonicum* is various from 2.55 to 2.65, while *C. elatius* with the relative density of 2.68–2.70, and 2.65–2.70 for *C. rubrum*.

### 3.4. Morphology features

The structures of most precious corals are typically consisting of two patterns. The first is ribbed or striated pattern that extends roughly parallel to the length of the coral branch. The other is a concentric, scalloped structure. Paralleled grooves are typically appear on the surface of *C. elatius* and *C. rubrum* (**Figures 10** and **11**), while the surfaces are relatively smooth on *C. japonicum* (**Figure 12**). In addition, natural dotting the coral surface, which may be described as pits and pockmarks, is only observed on *C. rubrum*. However, the paralleled stripes exist in the internal vertical section of all three species, no matter how different they look like on the surface (**Figures 13** and **14**).

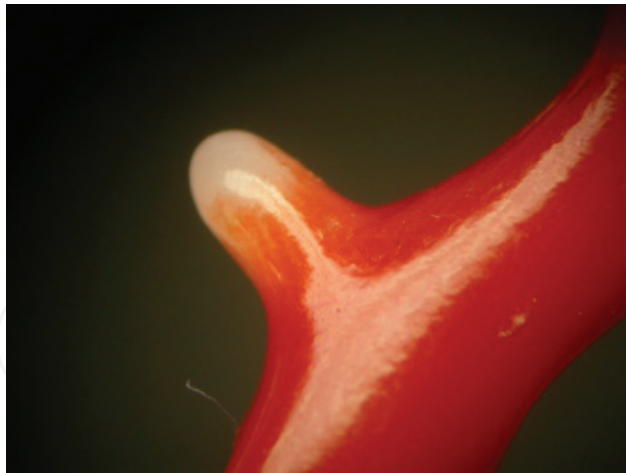
These two patterns are easily to be understood by recognizing the formation of polyps in precious coral. Actually, the longitudinal section of precious coral is made in correspondence with the oral disc at the bottom.



**Figure 10.** Paralleled grooves appear on the surface of *C. elatius*.



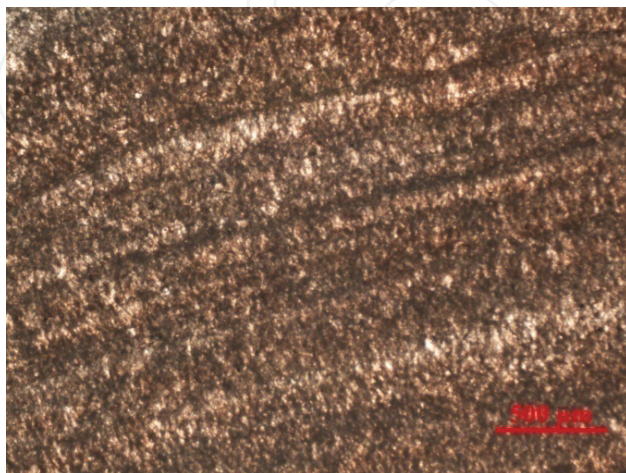
**Figure 11.** Paralleled grooves and pit marks appear on the surface of *C. rubrum*.



**Figure 12.** Relatively smooth surface of *C. japonicum*.



**Figure 13.** Micrograph of the cross section of precious coral.



**Figure 14.** Micrograph of the vertical section of precious coral.



4. General identification

Precious coral with its unique appearance and distinctive structure is easy to distinguish them from the common imitations such as dyed bone artifacts, dyed shell, dyed marble, conch pearl, Gilson coral, red glass, red plastic, and dyed wood. The identification features are listed in **Table 4**.

Among all these imitations, dyed corals are the most complicated. Some of the dyed corals are sharing the similar features on cross and vertical sections. Fan [6] acquired four specimens covering *C. japonicum*, *C. elatius*, *C. rubrum*, and *Isis hippuris*, which are the common species coral in trade market.

*I. hippuris*, also known as the name “bamboo coral”, is a white coral belonging to family Isididae, phylum Cnidaria, class Anthozoa, and subclass Octocarallia (**Figures 15 and 16**). The chemical composition and mineral component are keeping the same as the precious coral in Coralliidae family. It is also hard to tell the distinction from the texture and structure. As a result, dyed *I. hippuris* is always selected to be the material to imitate the precious coral (**Figures 17 and 18**). All the specimens are tested by laser Raman spectroscopy. The result

Properties varieties	Color	Transparency	Luster	Refractive index	Relative density	Moh’s hardness	Fracture	Others
Precious coral	White, pink-to-red	Opaque to semitransparent	Oily luster	1.48–1.65	2.70 (±0.05)	3–4	Even fracture	Color distribution is naturally uneven; concentric annulus on cross section; parallel stripes on vertical section acid blistering
Gilson coral	Red	Opaque	Waxy luster	1.48–1.65	2.44	3.5–4	Even fracture	No chromatic difference, fine grained texture; acid blistering
Dyed bone	Red	Opaque	Waxy luster	1.54	1.70–1.95	2.5	Splintery fracture	Uneven color on the surface and inside; round hole structure
Dyed marble	Red	Opaque	Glass luster	1.48–1.65	2.70 (±0.05)	3	Uneven fracture	Fine grained texture; acid blistering
Red plastic	Red	Transparent to opaque	Waxy luster	1.49–1.67	1.4	<3	Even fracture	Hot needle test (acrid odor); bubble inclusion
Red glass	Red	Transparent to opaque	Glass luster	1.635	3.69	5.5	Conchoidal fracture	Bubble inclusion
Dyed coral	Red	Transparent to opaque	Waxy	1.48–1.65	2.70 (±0.05)	3–4	Even fracture	Acetone reaction (color developing effect)
Conch pearl	Light salmon	Opaque	Waxy	1.486–1.658	2.85	3.5	Splintery fracture	Flame structure; acid blistering

**Table 4.** Identification features for precious coral and its imitations.





**Figure 15.** Raw *I. hippuris*.



**Figure 16.** Polished *I. hippuris*.



**Figure 17.** Dyed and polished *I. hippuris* (claret-colored).



**Figure 18.** Dyed and polished *I. hippuris* (scarlet color).

shows that the white part of the precious coral and *I. hippuris* has the same and distinctive spectrum of calcite. The interesting thing is that the red part of all precious corals shows a suit of peaks at 1016, 1128, 1296, 1518, 2147, 2250, 2633, 3032, 3361, and 3470  $\text{cm}^{-1}$ , while the dyed red *I. hippuris* has no featured peaks within this range. The study indicates that laser Raman spectroscopy is the useful, rapid, and nondestructive identification methods to distinguish precious coral from dyed *I. hippuris*.

## 5. Determination on the origin of precious coral's color

In the end of last century, carotenoids are determined to be responsible for a broad range of colors of plants and animals depending on the complex form and its incorporation into the host [7]. Rolandi et al. [8] attributed that various carotenoids are responsible for yellow, orange, brown, and blue-to-violet hues in coral skeletons. Kaczorowska et al. [9] opined that color of the red coral is caused by partial degradation and “leakage” of organic matter including beta-carotene from plant and algae material.

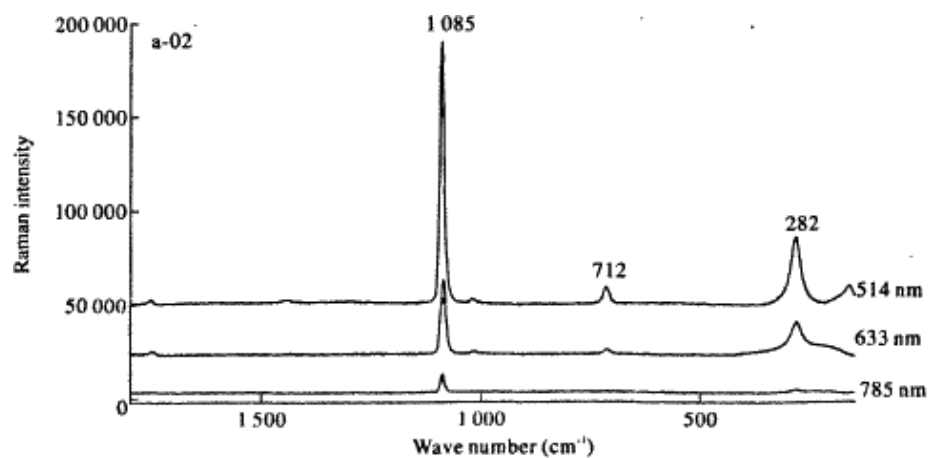
Raman effect arose from the inelastic collision between light and the molecule. The number, intensity, and shape of the Raman spectrum are correlative with molecular vibration or base group vibration. Raman scattering effect is a unique tool for the in situ study of biomineral [10]. Some previous studies using Raman spectroscopy have suggested that this method is useful to detect pigments in red corals. Understanding the nature of the pigments in the natural red corals can help to separate them from their equivalents. The present study shows that precious corals may contain more than one pigment.

This study was carried out on various coral samples of *C. japonicum*, *C. elatius*, and *C. rubrum*. Samples of *C. japonicum* and *C. elatius* are selected the ones with white portions. Samples were tested by 514, 633, and 785 nm laser of Renishaw inVia micro-Raman spectrometer at normal temperature and pressure. The power on the sample was 2 mW, whereas the acquisition time

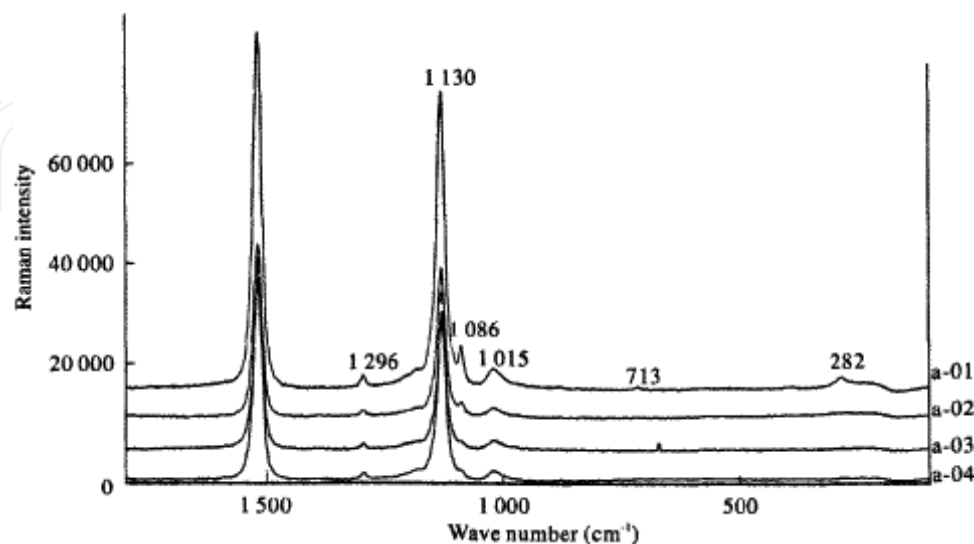
was 20 s and the slits were set at 40  $\mu\text{m}$ . Raman tests were carried on at red color zone and white color zone of the specimens for the detection of chromatic composition. PeakFit 4.12 software was adopted to overlap the peaks at approximately  $1500\text{ cm}^{-1}$  for finer analysis.

Not surprisingly, all the specimens present the diagnostic peaks of calcite at  $1085\text{ cm}^{-1}$  among  $100\text{--}1800\text{ cm}^{-1}$  range. In **Figure 19**, Raman spectra of a white portion in *C. japonicum* and *C. elatius* using three different excitation wavelengths are shown. Raman peaks are due to calcite at  $1085\text{ cm}^{-1}$  ( $\nu_1$ , symmetric mode of carbonate),  $712\text{ cm}^{-1}$  (in-plane bending mode of the carbonate), and  $282\text{ cm}^{-1}$  (crystal lattice vibration of calcite).

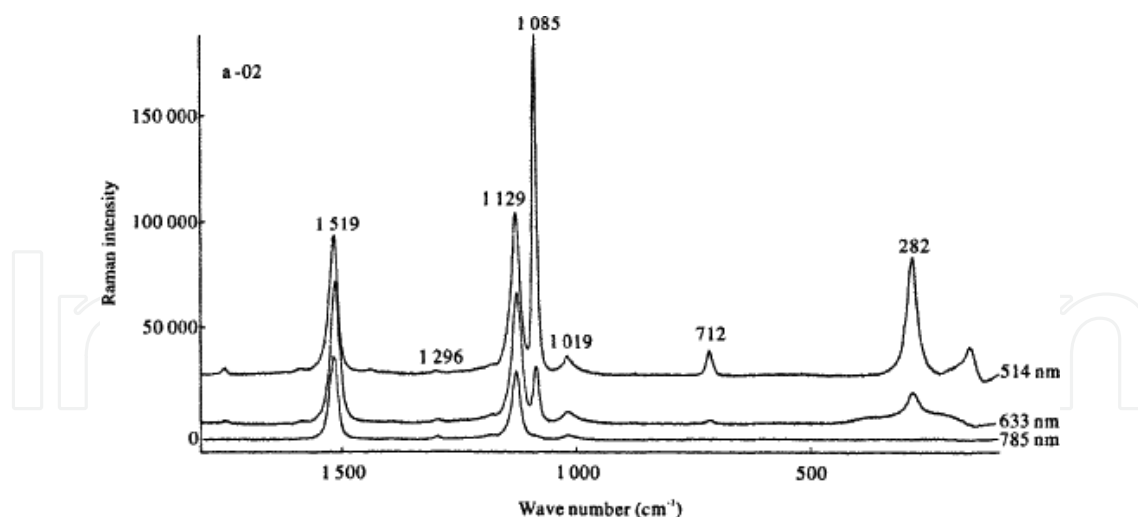
In **Figure 20**, Raman spectra of four different precious coral samples, using the 514 nm excitation, show two intense additional bands in the region  $1000\text{--}1800\text{ cm}^{-1}$  compared to the spectra of white core in samples. In all the samples tested in this study, these peaks can be found. It is clear that these peaks, which are characteristics of polyenic chains, are responsible for the pigmentation in the samples.



**Figure 19.** Raman spectra of the white core in the sample, using three different excitation wavelengths.



**Figure 20.** Raman spectra of the red parts in the samples, using 514 nm excitation wavelength.



**Figure 21.** Raman spectra of the red parts in the samples, using three excitation wavelengths.

**Figure 18** shows the Raman spectra of samples presented in **Figure 21**, which is measured by three different excitation wavelengths. In these figures, changes in shape and relative intensities occur at the same point by changing the excitation wavelength. Region  $1500\text{ cm}^{-1}$  is sensitive to the length of the polyenic chain.

All the samples show the two major Raman peaks at  $1130$  and  $1519\text{ cm}^{-1}$ . These peaks were not detected in the white core of the precious coral samples.  $\text{C}=\text{C}$  ( $\nu_1$  near  $1500\text{ cm}^{-1}$ ) and  $\text{C}-\text{C}$  ( $\nu_2$  near  $1130\text{ cm}^{-1}$ ) stretching vibration observed only in red coral samples suggests that these compounds play the main role with regard to the color. The series of bands observed by decomposition can be assigned to a series of polyenic molecules lacking methyl groups. Different colors of the precious corals are because of different mixtures of pigments in varying relative proportions. These kinds of pigments are not found free in nature but rather are complexed with some forms of a protein. A  $\text{CaCO}_3$  – pigment complex has been proposed for the calcareous skeletons of corals and some shells. In the absence of an adequate protein complex, the pigments in the precious corals could possibly be complexed with carbonates. All of the samples are colored by a mixture of pigments. Different colors are explained by different mixtures, not by a single pigment. As we mentioned in Section 3.1, organic composition, even is a small amount, plays an important role on the color of precious corals.

## 6. Determination on organic matrix and biogenic calcite in precious corals

In the process of determining the origin of precious corals' color, we are very clear about the existence of organic matter and its role. In spite of detailed studies, the internal structure of the axial skeleton of precious corals is not understood. In particular, the spatial relation between the organic matrix and biogenic calcite remains in great part unexplored. Curiously, the answers for how is the spatial distribution of the organic matter, the interface between the organic matter and biogenic calcite, the effect of the organic matter produced during the mineral construction of precious corals are still need to be studied.

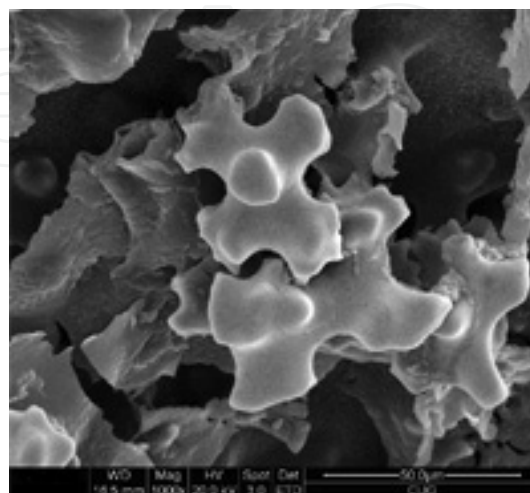
Different techniques, such as SEM and HR TEM, will be applied to study how each hierarchical layer of precious corals assembles into larger units. EBSD and TEM studies will show the degree of crystallographic misorientation between the building blocks. The approach of in situ analysis will be carried out by means of sputtering ion, decalcification, and structural analysis techniques. The method is to observe the spatial distribution of organic framework and organic/inorganic interface spatial relationship in multilevel. The study is to understand a multiscale physico-chemistry characterization of mineral part of precious coral and its three-dimensional architecture.

### 6.1. Organic matrix

Since the amount of organic matter is very slight in precious coral, an inclined decalcification experiment was conducted in the study. First, fine polished vertical and cross sections of precious coral samples were fixed on an inclined specimen holder. Then EDTA (Ethylenediaminetetraacetic acid disodium salt) solution in certain concentration was dropping from dropper to etch the surface of the samples gradually. Several hours later, a transition surface including fine polished part, semi-etched part, and fully decalcified part was formed. Spicules and organic matrix of precious coral were observed by SEM.

We observed several types of spicules, such as cross spicule (**Figure 22**), eight-axial spicule (**Figure 23**), six-axial spicule (**Figure 24**), and double eggplants spicule (**Figure 25**). Organic matrix could be easily found around the surface of the spicules.

The organic matrix net is distributed in layers to form a three-dimensional cavity (**Figure 26**), which provides the growth space for the spicule. Most of spicules were wrapped by organic matrix (**Figure 27**). But, we also observed some flaw parts of the samples (**Figures 28 and 29**) lacked of organic matter led a sprawling growth habit, which indicate the growth of precious coral were under the control of organic matter.



**Figure 22.** Cross spicule.



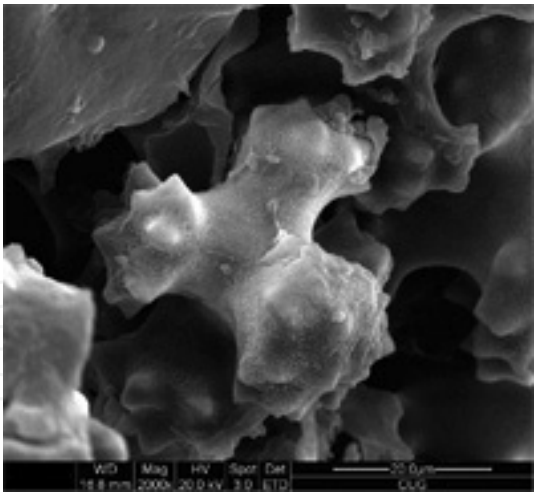


Figure 23. Eight-axial spicule.

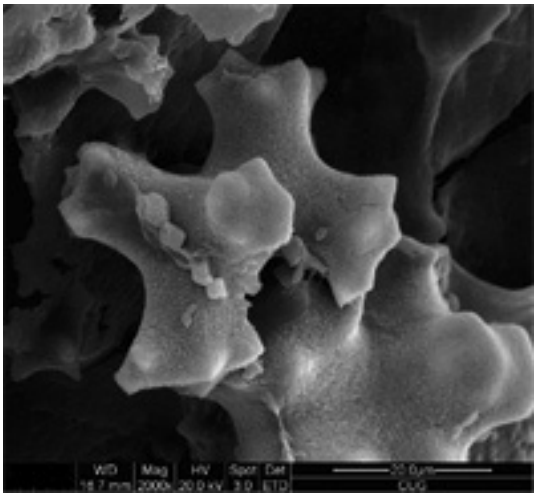


Figure 24. Six-axial spicule.

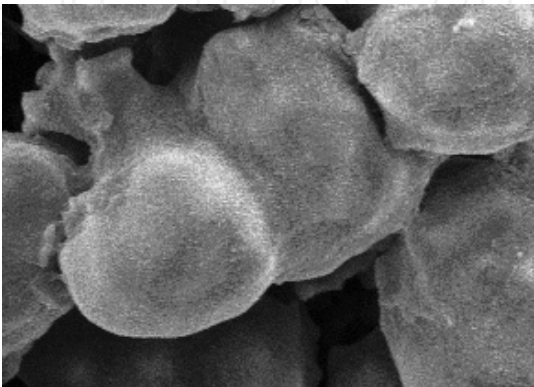
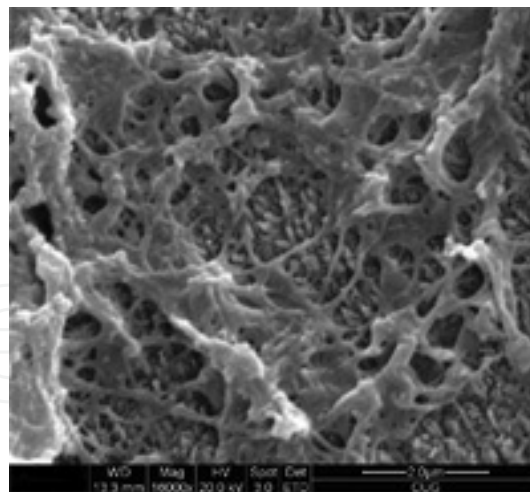


Figure 25. Double eggplants spicule.



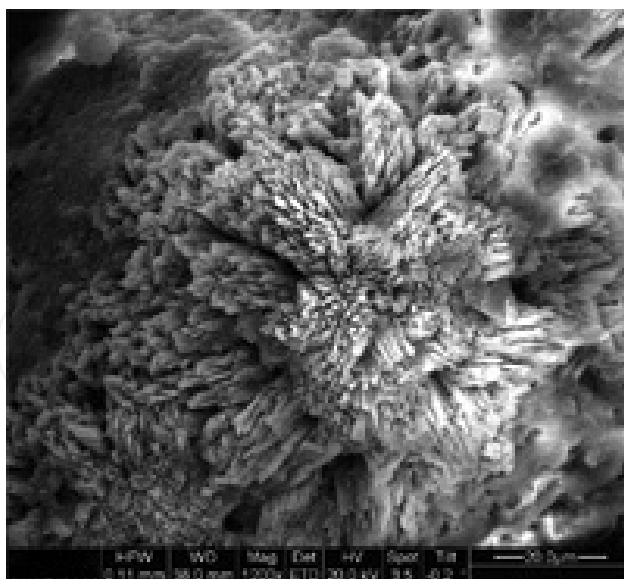
**Figure 26.** The organic matrix net is distributed in layers to form a three-dimensional cavity.



**Figure 27.** Spicules were wrapped by organic matrix.



**Figure 28.** Sample with flaws.

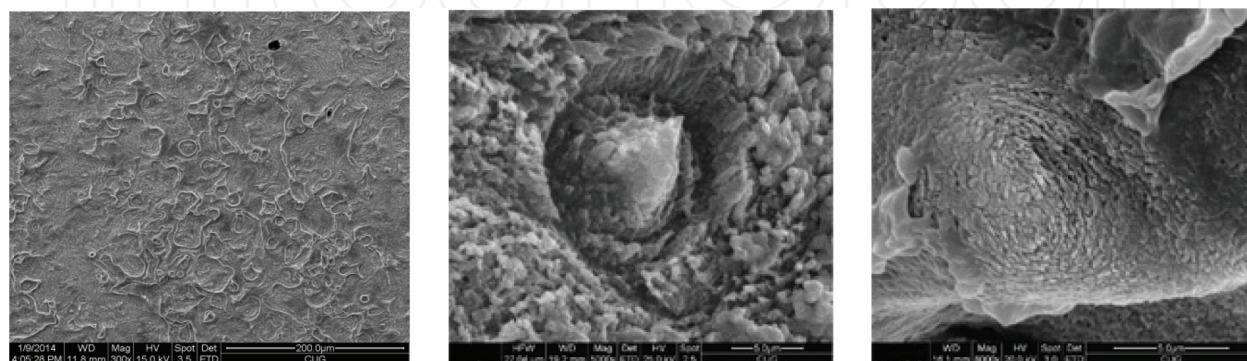


**Figure 29.** Radial growth of calcite crystal without the control of organic matter.

## 6.2. Multilevel modular mesocrystalline organization in precious coral

Biomaterials often display morphological, chemical, and crystallographic patterns at length scales ranging from the nanoscale to the macroscale (**Figure 30**).

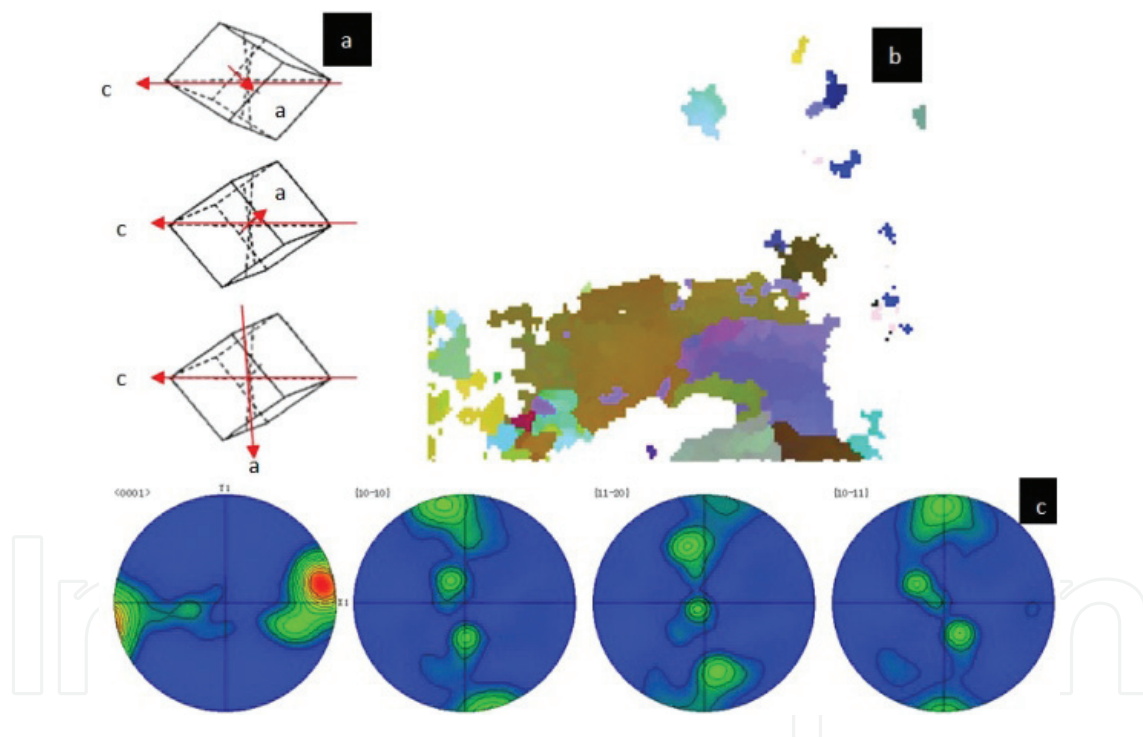
In the skeleton of precious coral, we observed that building blocks were arranged into several hierarchical levels of oriented modules. The modules in each hierarchical level assemble into larger unit that comprises the next higher level of the hierarchy. The EBSD and TEM studies show the degree of crystallographic misorientation between the building blocks, which decreases with decreasing module size (**Figure 31**). The crystal units of axial skeleton are columnar with two perpendicular directions. Throughout the region, the orientation of c axis is consistent with its long axis of each unit. The orientation of a axis and b axis of each unit is relatively complex with four kinds of orientation. One orientation is parallel to the plane. The other orientation is perpendicular to the plane. The remained orientation crosses the plane



**Figure 30.** Precious coral skeleton formed by nanoscale to the macroscale calcite crystals.



**Figure 31.** TEM test shows the crystallographic orientations of precious coral sample.



**Figure 32.** The misorientation angle of different units of precious coral is between 33 and 48°. The misorientation angle is nearly 5° in the area of similar orientation by EBSD test.

with different skew angles. The misorientation angle of different units is between 33 and 48°. The misorientation angle is nearly 5° in the area of similar orientation (**Figure 32**).

That is to say, precious coral, as a member of biomineral family, assembled skeleton as a delicate arrangement of a hierarchy of crystals with well-defined orientations under the control of organic matrix.

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