論文の内容の要旨

Evolution and diversity of biomineralogical nature in ectocochleate cephalopod septa

（外殻性有殻頭足類の隔壁における結晶学的性質の進化と多様性）

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Septa, characteristic structures of ectocochleate cephalopods, enable ammonoids and nautiloids to gain buoyancy and swimming ability by restoring low-pressure gas in the phragmocone. The macro-morphological differences in septa between ammonoids and nautiloids have been intensively examined, and their functionality and morphogenetic mechanisms have been well discussed. However, little attention has been paid to their biomineralogical nature; the septa in these two groups are regarded to be biomineralogically homologous. Determination of biomineralogical differences is essential for recognizing the functional morphology and biomineralization process of structures unique to cephalopods. This study conducted comparative crystallographic analyses on the nacreous structure of ectocochleate cephalopod septa. The crystallographic orientation pattern, (crystallographic texture) and microstructure (morphology and mode of layering of the constituent polycrystalline materials) of the septa were analyzed and compared with a focus on the effects by phylogeny and habitat environments.

In Chapter 1, as an introduction, macro-morphological diversity of ectocochleate cephalopod septa and its functional morphology are reviewed. Then, the significance of biomineralogical properties for the elucidation of functional morphology of septa is mentioned.

In the study described in Chapter 2, the septa of extant nautiloid individuals such as *Nautilus pompilius*, *N. belauensis*, *N. macromphalus*, and *Allonautilus scrobiculatus* from various geographic localities including the Philippines, Fiji, Palau, New Caledonia, and Indonesia in addition to those raised in an aquarium were analyzed by using X-ray diffraction (XRD) to determine their crystallographic textures. The {002}, {012}, {102}, {112} pole figures generated from the diffracted pattern reveals that, in general, the septa are composed of aragonitic polycrystals with their *c*-, *a*-, and *b*-axes perpendicular to the septal surface, parallel to the dorsoventral direction, and parallel to the left-right direction, respectively. These characteristics indicate an ordered texture. However, deviations from the ordered texture also exist in the *a*- and *b*-axes directions, showing a slightly disordered texture. The degree of deviation was measured on the basis of the full width half maximum (FWHM) values calculated from the {012} pole figures. Combining the *c*-axis orientation indicated by {002} pole figures, two FWHM values in the {012} pole figures were utilized as indices of *a*- and *b*-axes misalignment. As a result, variability in *a*- and *b*-axes misalignment in septal aragonitic polycrystals among individuals was revealed. They were constant in each septum and submature–matured septa in each individual. Moreover, individuals from the same phylogeographic group shared similar FWHM values. In this study, the relationship of larger FWHM values with a markedly disordered crystallographic texture and a shallower-habitat phylogenetic group is reported for the first time. The investigation on aquarium-raised individuals confirmed that sea water pressure does not abiotically affect the crystallographic texture. These results suggest that the crystallographic texture of the recent nautilid septa are influenced genetically and reflect certain adaptive significance toward their habitat water depth.

In the study described in Chapter 3, the microstructural analyses on recent nautilid septa were conducted qualitatively and quantitatively. The nautilid septa are known to be composed entirely of nacreous structures. In this study, the morphology and mode of layering of the tablets that construct the nacreous structure were examined and measured by using scanning electron microscopy and synchrotron X-ray nano computed tomography. As a result, the recent nautilid septal nacre is characterized by a laterally long, thin, and elongated hexagonal shape. These tablets are stacked in a columnar fashion in a broad view as reported in previous studies. However, the columns are not continuous throughout the septa; rather, they are often separated in two columns that in some cases fuse into one column. Observations of the concave septal surface revealed that such a mode of layering is achieved by tablets growing on the margins of the underlying tablets. In addition, the elongated hexagonal tablets are well aligned with each other, and “twinning” is often present. Along with the crystallographic texture investigated in Chapter 1, the *a*-axis of the nacreous tablet is parallel to the elongated direction of the tablets. The nautilus septal nacre is similar to that of pinnid and nuculid bivalves, which has been previously described as a row-stack nacreous structure. Because the nautilus septal nacre is regarded as a having a columnar nacreous structure, which is also observed in gastropod nacre, this finding is important in comparative studies of molluscan nacreous structures. The measurements of nacreous tablets revealed no distinct variation in the septal nacre of recent nautilus belonging to different phylogeographic groups.

In Chapter 4, fossil ectocochleate cephalopod septa were compared with those of recent nautilids examined in the previous chapters. Co-occurring species of ammonoids and nautiloids with similar habitat depth were utilized to exclude the influence of habitat environment on the septal biomineralogical properties. The examined specimens include the Upper Cretaceous nautiloid *Eutrephoceras*, scaphitid ammonoid *Hoploscaphites nodosus*, and *H. brevis* from the western interior province of North America in addition to the Lower Cretaceous nautiloid *Cymatoceras* sp. and douvilleiceratid *Douvilleiceras* sp. from Madagascar. The fossil nautiloids show crystallographic textures and microstructures similar to those in recent samples, i.e., *a*-axis parallel to the vertical direction and *b*-axis parallel to the left-right direction, although the degree of misalignment was greater in the fossils. The sizes of the tablets in the fossil nautiloids are within the same range as those of the recent specimens. However, the tablet stacks clearly show a columnar fashion. The examined fossil species are considered to have had shallower habitats. Considering the fossil record of nautiloids, it is suggested from a biomineralogical perspective that the septa and their formation process have been conservative since the early Mesozoic; however, the crystallographic misalignment and mode of layering of the nacreous tablets might reflect their variable habitat depth. In contrast, the ammonoid septal nacre shows polygonal tablets with thicknesses similar to those of the nautiloids. Moreover, although their crystallographic texture shows a *c*-axis perpendicular to the septal surface, as in the nautiloids, it is completely disordered in *a*- and *b*-axes, which are represented by ring-like pole figures in the {012}, {102}, and {112} planes. This result suggests that the crystallographic properties of ectocochleate septa, including their crystallographic orientation and nacreous structure, are conservative at the subclass level. However, they might vary considerably between the Ammonoidea and the Nautiloidea, which developed different septal morphologies.

In Chapter 5, these new insights are discussed for understanding the evolution of ectocochleate septa, particularly from aspects of morphogenesis and functional morphology. The similarity of the crystallographic properties of nautiloid septa to the bivalves suggests a terrace-like formation of nacre. The alignment of tablet elongation and crystallographic ordering in bivalves are explained by geometric selection based on the faster growth along in the *b*-axis of aragonite than that along in the *a*-axis. Such crystal growth due to the geometric selection might occur in nautiloid septal nacre, because their nacreous tablets are also elongated. However, the elongation direction and crystallographic orientation is inconsistent with those of bivalves. The mechanisms for the vertical stacking of the nautiloid septal nacreous tablet through the interlamellar membranes would be similar to those of bivalves because the similar mode of tablet layering indicates the existence of mineral bridges produced by rupture owing to permeability pressure.

Contrary to the bivalve-like nautiloid nacre, the ammonoid columnar nacreous structure, which is similar to the "stack of coins" structure in gastropods, can be considered to have formed by mineral bridges located in the central area of the tablets. The completely disordered crystallographic orientation pattern of the ammonoid septal nacre is consistent with such a gastropod-like nacre growth mechanism, suggesting that weak or no competition/interaction with lateral adjacent tablets/columns. These differences in formation mechanisms for nautiloid and ammonoid septa indicate that different morphogenetic constraints might exist in their septal formations, which affect the macro-morphological difference of septa, based on the different properties and dynamics of the septal mantle and the septal mantle epithelium.

The nacreous structure is known to be mechanically excellent among molluscan shell microstructures. Therefore, the development of the nacreous structure in septa would be reasonable to resist high pressure in the ambient surrounding water because low-pressure gas fills the phragmocone. However, whether or not the nacreous structure is mechanically optimized and performs best depends on the constituent tablet size and its mode of layering. In this respect, the mechanical performances and optimization of both nacreous tablets, particularly the lateral and overlap length, were compared by using a multi-objective optimization model. The results indicate that the tablet and overlap aspect ratios of the nautiloids are close to the optimal values to perform high tensile strength, stiffness, and toughness. On the contrary, the septal nacreous tablets of the ammonoids are short in lateral and overlap lengths toward the thickness, suggesting that they are not optimized in these mechanical properties. This thesis revealed not only the difference in the mechanical properties of septa, but also the anisotropic characteristics of septa, especially in nautiloids. Such anisotropy of the septa necessitates reconsideration of its functional macro-morphology, because previous numerical analyses on the septal macro-morphology have been assumed the isotropy of the septal material. This study demonstrates for the first time, that the biominelogical studies on ectocochleate cephalopod septa is important not only for biomineral research itself, but also important to understand the evolution and ecology of macro-morphology of septa.

This thesis suggests that the gastropod- and bivalve-type nacre formation mechanisms themselves are not different because the both types are present in the ectocochleate cephalopod septa. The septa are flexible and continuous according to their habitat environments, although the genetic and evolutionary origin might not be homologous among mollusks. Moreover, it is suggested that extremely subtle differences in the crystallographic structure in genus, species, and individual levels should be considered.