An Examination of the Micro-Crystals of Calcium Carbonate in Molluscan Shells by Means of X-Rays, Part III

By

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Abstract

Testing the shells of more than 60 species of Japanese Molluscs by means of the X-ray method, it is proved that there exist various degrees of fibrous arrangement of the micro crystals of calcium carbonate in the molluscan shells. In all of them the axes of the fibrous arrangement are in a direction nearly perpendicular to the surface of the shell, coinciding with the principal axis of calcite or with the orthorhombic *c* axis of aragonite respectively.

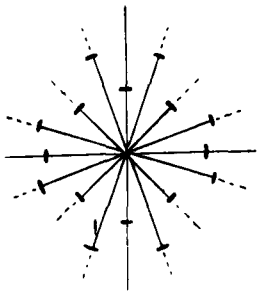
The form and arrangement of the micro-crystals of calcium carbonate in *Atrina*, *Avicula*, *etc.* were examined by Ebner, Biedermann and others[1] nearly thirty years ago with the microscope or by passing a plainly polarized light through the shells. Such optical methods, however, are only available in a few kinds of shells which are composed of sufficiently large micro-crystals of calcium carbonate. For the majority of the shells they are inefficient for that purpose, whereas, the X-ray method involves no such restriction, and is good for any kinds of shells. As was stated in his previous papers[2], the writer utilized the monochromatic X-rays from the

1. Biedermann, W.; Ztschr. f. all. Physiol., Bd. **1**, (1902)
   „ „ ; Jena Ztschr., Bd. **36**, (1901)
   „ „ ; Biol. Ctbl., Bd, **21**, (1901)
   Ebner, V. ; Sitz.-ber. d.Wiener Akad., math.-nat. Kl., Abt. 2, Bd. **89**, (1884–85) & Bd. **91**, (1885)
   Dippel, L. ; Zeitschr. f. Wiss. Mikrosk. Bd. **17**, (1900)
2. J. Tsutsumi ; These Memoirs, A, **11**, 217 & 401 (1928)

Mo-target in the examination of the micro-crystals of calcium carbonate in the shell of *Atrina japonica* Reeve, and was able to determine their crystal-forms as well as their arrangement very easily. The same method was applied now to examine various shell specimens of more than 60 different species of Molluscs. The results thus obtained are as described below.

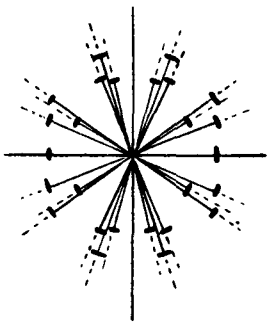
A diagram of the X-ray diffraction patterns obtained with the prismatic and the nacreous layers respectively of the shell of *Atrina japonica* Reeve, by a parallel incidence of the X-rays to the surface of the layer, are shown here again in Text-figs. 1 and 2. Fig. 1[1] illustrates the

Fig. 1



The prismatic layer

Fig. 2



The nacreous layer

X-ray diffraction pattern of the prismatic layer, most parts of which are composed of micro-crystals of calcite, while Fig. 2[2] shows that of the nacreous layer, most parts of which are composed of micro-crystals of aragonite. The revelation of the diffraction patterns as shown in these figures indicates that the micro-crystals of calcium carbonate in both these layers are arranged fibrously, and also that the axis of such fibrous arrangement is nearly parallel to a direction perpendicular to the surface of the layer coinciding with the principal axis of calcite or the orthorhombic *c* axis of aragonite respectively.

First the diffraction photographs with X-rays were taken of about 10 different shells belonging to the order of Lamellibranchia, by sending the X-rays in a direction parallel to the surface of the layer of the shells, and some of them have been reproduced in Plate I, Figs. 1, 2, 3 and 4 as

1. J. Tsutsumi ; These Memoirs, A, **11**, 217 (1928) & H. Fukushima ; „ „ A, **11**, 223 (1928)
2. J. Tsutsumi ; „ „ A, **11**, 401 (1928)

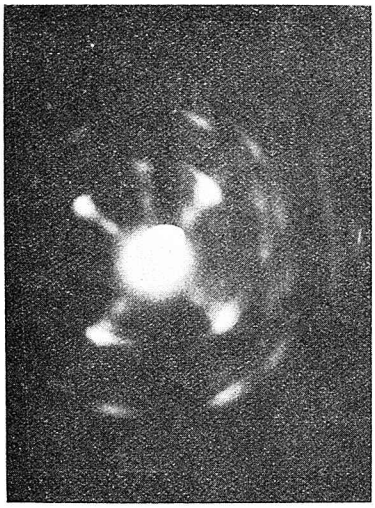
typical ones. Figs. 1 and 2 in Plate I are diffraction figures obtained with a shell of *Mytilus crassitesta* Lischke. This shell consists of two layers, each of which is stratiform. In the outer layer of the shell the calcium carbonate is in the form of calcite, while that in the inner layer is in the form of aragonite. By comparing these photographs with the foregoing Text-figs. 1 and 2, we see that the micro-crystals of calcium carbonate in the shell are arranged fibrously in the direction nearly perpendicular to the surfaces of the layers, and that the principal axis of calcite as well as the orthorhombic *c* axis of aragonite are arranged as the axis of the fibre, as was the case in the prismatic or in the nacreous layers of *Atrina japonica* Reeve. The diffraction patterns obtained with *Anomia lischkei* Dautzenberg et Fischer and *Pinctada martensi* Dunker, which also belong to the order of Lamellibranchia, are reproduced in Plate I, Figs. 3 and 4. The distribution of radiating bands and the dispersal of intense spots on these bands perceived in these photographs are essentially the same as in Figs. 1 and 2 in Plate I respectively. This fact indicates that the crystal-forms and the arrangement of the micro-crystals of calcium carbonate in the shell of *Anomia* and *Pinctada* are identical respectively with those of the micro-crystals in the prismatic and in the nacreous layers of the shell of *Atrina japonica* Reeve. Next the writer examined the shell of 8 more different species in the same order and was able to ascertain that the crystal-forms and the arrangement in question are the same as those in the nacreous layer of *Atrina japonica* Reeve. Then the writer tried different shells of ca. 30 species of Eulamellibranchia, 4 species of Aspidobranchia, 6 species of Pectimibranchia, 2 species of Streptoneura, 1 species of Placophora, and 1 species of Dibranchia. The diffraction patterns obtained with the shells of *Sunetta excavata* Hanley, *Gomphina melanaegis* Roemer, *Turbo cornutus* Solander, *Fusinus perplexus* A. Adams, *Haliotis asinina* Linne, *Cardium muticum* Reeve, *Dentalium hexagonum* Gould are shown, as typical ones, in Figs. 5, 6, 7, 8, 9, 10 and 11 of Plates I and II. All these photographs were taken by setting the surface layer of the shells parallel to the X-ray beam. The results thus obtained with most of the shells are again essentially the same as in the case of the nacreous layer of *Atrina japonica* Reeve. There are, however, some exceptional cases; in *Fusinus perplexus* A. Adams, *Dentalium hexagonum* Gould, most of the micro-crystals are not arraged fibrously. Moreover in *Cardium muticum* Reeve the micro-crystals are arranged quite irregularly as seen in Plate II, Fig. 10, which is a photograph taken by setting the shell of *Cardium* in various orientations; i. e. to be exposed to the incident X- ray beam.

Up to now the writer has examined the shells of more than 60 different species belonging to various orders, arriving at the following general conclusion. Calcium carbonate in the porcelaneous and the nacreous layer of the molluscan shells is in the form of aragonite while that in the prismatic layer of them is usually in the form of calcite, but in a few cases in the form of aragonite. As to the constitution of the micro-crystals of calcium carbonate, there are various degrees of the fibrous arrangement, but the axis of the fibrous arrangement is always in a direction nearly perpendicular to the outer surface of the shell, and coincides with the principal axis of calcite or the orthorhombic *c* axis of aragonite respectively. So far as the writer's experiment goes, it seems that the more perfect the appearance of the strata, the more perfect the fibrous arrangement of the micro-crystals of calcium carbonate. Since the shell materials for this test was so selected as to cover as evenly as possible all orders of Molluscs, it seems to the writer reasonable that the general conclusions stated above are also good for the other shells not yet examined.

This study was carried out under the supervision of Prof. T. Kawamura and Prof. U. Yoshida, to whom the writer's sincere thanks are here expressed. He is also indebted to Mr. T. Kuroda who was so kind as to assist him in the present investigation.

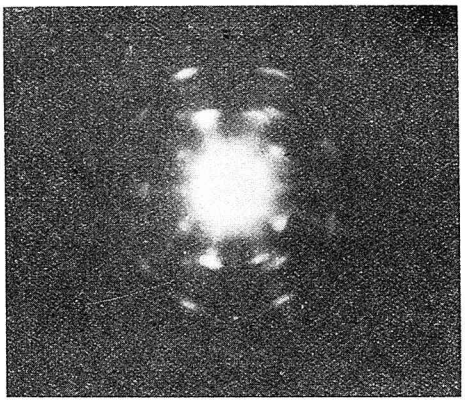
Plate I

Fig. 1



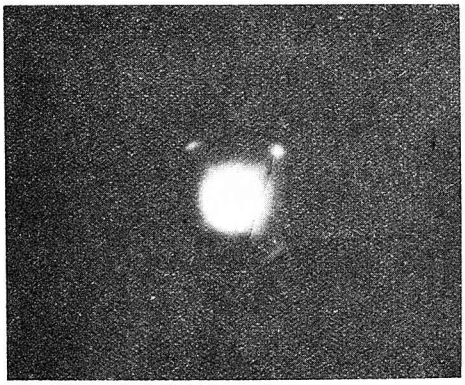
The outer layer of
Mytilus crassitesta Lischke

Fig. 2



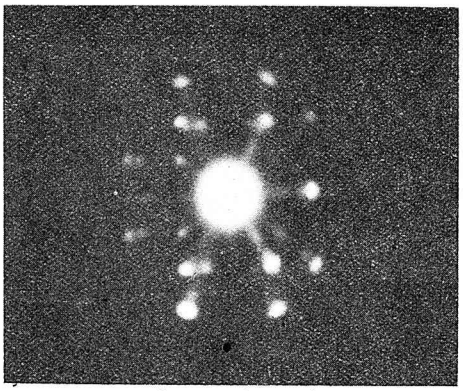
The inner layer of
Mytilus crassitesta Lischke

Fig. 3



Anomia lischkei
Dautzenberg et Fischer

Fig. 4

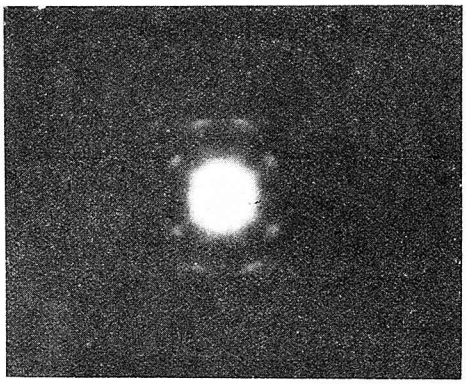


The inner layer of
Pinctada martensi Dunker

Fig. 5

Sunetta excavata Hanley

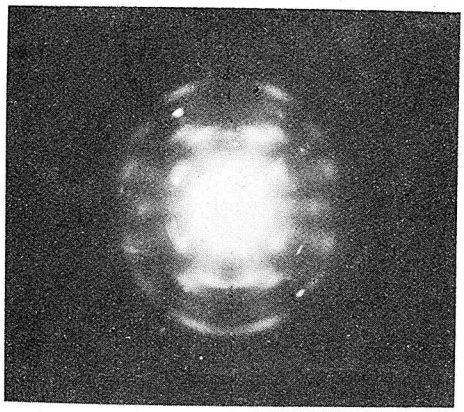
Fig. 6



Gomphina melanaegis Roemer

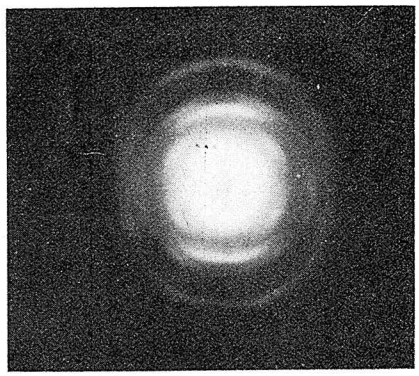
Plate II

Fig.7



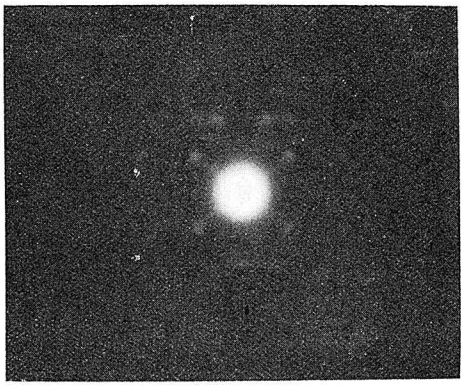
The inner layer of
Turubo cornutus Solander

Fig.8



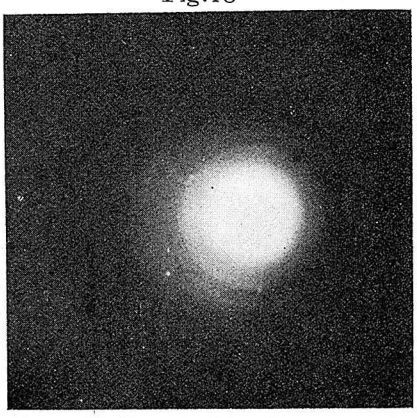
Fusinus perplexus A. Adams

Fig.9



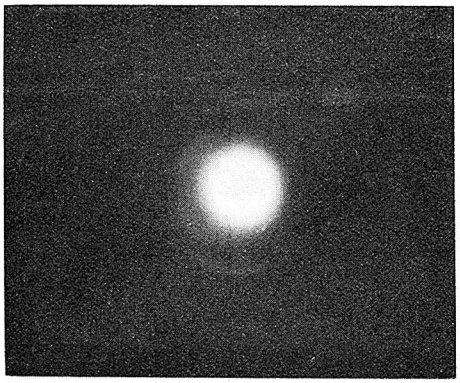
Haliotis asinina Linne

Fig.10



Cardium muticum Reeve

Fig.11



Dentalium hexagonum Gould