16. NEW DATA ON THE MOLECULAR SYMMETRY AND ORGANIZATION OF THE QUASI-CRYSTALLOID SKELETON OF THE SPORODERM

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Abstract

During our experimental investigations on the partially dissolved and degraded pollen grains of *Ambrosia artemisiifolia* L. a degraded regular pentagon biopolymer unit was investigated with two dimensional symmetry operations. The tenfold primary rotation method resulted cyclic molecular clusters on the places of the globular units of the quasi-crystalloid skeleton in Angstrom dimension. This is the first molecular data about the globular units forming the regular pentagon which are the building elements of the metastable quasi-crystalloid skeleton. In this contribution we present a short and selected review on the results of the biopolymer organization and symmetry of the sporopollenin.

Key words: Palynology, *Ambrosia*, recent, molecular structure.

Introduction

The publications of ROWLEY (1978), ROWLEY, J.R., DAHL, and ROWLEY, J.S. (1980), ROWLEY, J.R., DAHL, SENGUPTA and ROWLEY, J.S. (1981), ROWLEY, J.R., EL-GHAZALY and ROWLEY, J.S. (1987) and SOUTHWORTH (1985a,b, 1986a,b) started investigations with the TEM method the sub-units of the partially degraded exines of the pollen grains. Following the discovery of the quasi-crystals on rapidly cooled AlMn alloy by SHECHTMAN, BLECH, GRACIAS and CAHN (1984) intensive researches started in the crystallography and the symmetry of inorganic and organic structures.

In 1988 the quasi-crystalloid biopolymer structure was discovered from living systems, first from the exine of *Pinus griffithii* MCCLELL (KEDVES, 1988). Later this metastable biopolymer skeleton was investigated by several, different methods on several biological objects. These may be summarized as follows:

1. Two dimensional symmetry operations with the modified Markham rotation method (KEDVES, 1989a, 1990, 1991b, KEDVES and FARKAS, 1991, KEDVES, FARKAS, MÉSZÁROS, TÓTH and VÉR, 1991, etc.)
2. Three dimensional modelling for the metastable quasi-crystalloid skeleton (KEDVES, 1991a, 1992).
3. Two dimensional modelling for the stabilizing biopolymer system of the metastable skeletal elements (KEDVES and TÓTH, 1994).
4. Computer modelling for the skeletal and the stabilizing biopolymer systems (KEDVES, M. and KEDVES, L., 1995, 1996, 1997, 1999).

5. After the discovery of the peculiar biopolymer organization of the wall of *Botryococcus braunii* KÜTZ. isolated from Hungarian oil shale, namely there are quasi-crystalloid network and larger units which may be modelled with fullerenes (KEDVES, ROJIK and VÉR, 1991) a peculiar attempt was made to the relationships and connections of these two completely contradictory organization. Till this time the relationships of the quasi-periodic and quasi-equivalent systems in biological material is not yet resolved but the first partial results were published by KEDVES, TRIPATHI, VÉR, PÁRDUTZ and ROJIK (1998).

The metastable biopolymer skeleton and one kind of the hypothetical stabilizing units was compiled on the basis of the results of transmission electronmicroscopical data (KEDVES, 1989b). As it was emphasized in the above mentioned paper the Penrose tiling was re-discovered exclusively based on the TEM results of the partially degraded exines, because first the author has not read the literature of the Mathematics. The most important publications of PENROSE (1979), MACKAY (1976, 1981, 1990), BURSILL and PENG JU LIN (1985), SACHDEV and NELSON (1985), AUDIER and DUYOT (1986), NELSON (1986), O'HANDLEY (1987), SCHNEER (1988) and the book of HARGITTAI (1990) were studied later.

General problems

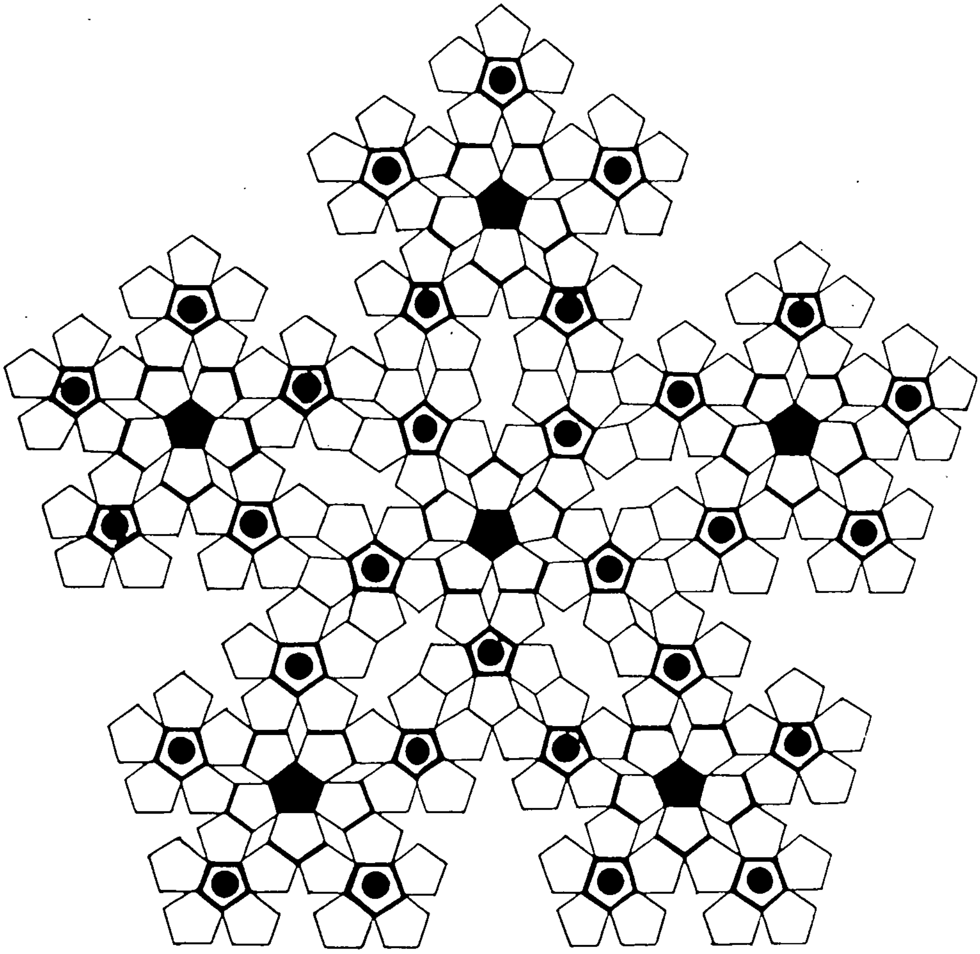
In the first place we re-publish the schema of KEDVES (1989b) concerning the metastable Penrose tiling (Text-fig. 16.1.). The most important characteristic is the presence of the frustrations sensu NELSON (1986). The quasiperiodic network does not fill the space, another so-called stabilizing system assure the equilibrum. On this schema one central stabilizing unit was illustrated. But without doubt there are several different kinds of molecular structures, which are filling the holes of the quasi-periodic network.

Text-fig. 16.2. is the reproduction of the fig. 1, p. 63 from the paper of KEDVES (1989b) illustrating the organization levels of the sporopollenin. The lower part is the molecular one sensu strictu. The molecular composition of the sporopollenin is also another very complicated problem, to this we cite without completeness some publications: BROOKS and SHAW (1973), MANSKAYA, KODINA and GENERALOVA (1973), RITTSCHER, GUBATZ and WIERMANN (1987), SCHULZE, OSTHOFF and WIERMANN (1987), KEDVES (1991b).

After the molecular system the units in Angstrom dimension are important. Two components are illustrated: 1. The metastable quasi-crystalloid skeleton. 2. The central stabilizing molecular system of the metastable skeleton. The most important characteristic feature of the units of this level of organization (diameter of the regular pentagon 14-28 Å about) is that after the Markham rotation of the regular pentagonal biopolymer units further points of symmetry appear, which are suitable for further symmetry operations, and among these the Penrose unit was also demonstrated. The quasi-crystalloid basic skeletal units may be building elements for further highly organized systems such as the helical, lamellar, globular structures in nanometer dimension.

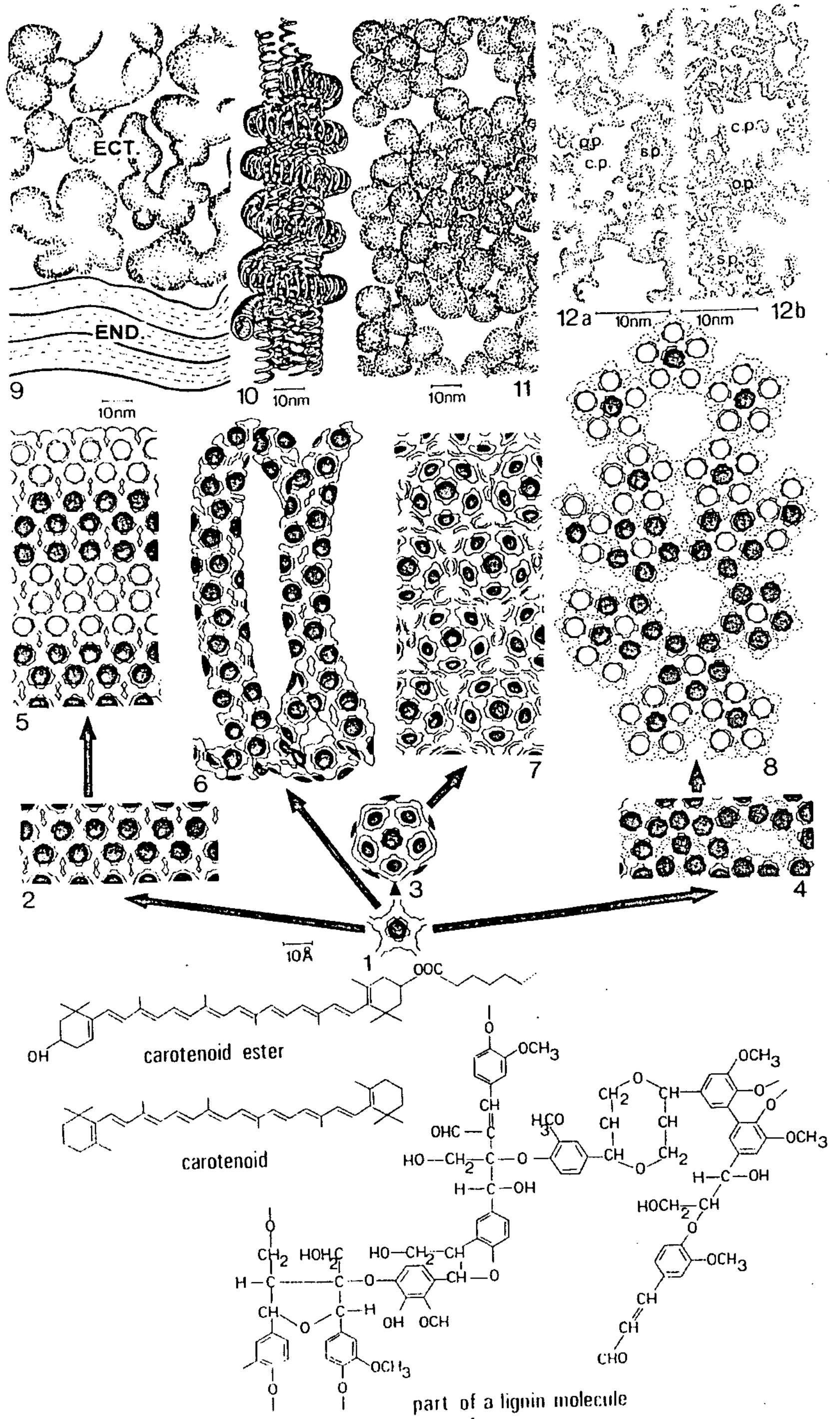
Regular pentagons may occur in nanometer dimension also (ROWLEY, 1967) but the Markham rotation never results in secondary points of symmetry.

Resuming the dimension is very important in the basic characteristic features of the regular pentagonal biopolymer structures, similarly to the colloids of the inorganic or organic materials.



Text-fig. 16.1.

Schema for the quasi-crystalloid biopolymer structure of the sporoderm, following KEDVES (1989b), p. 65.



Text-fig. 16.2.

The organization levels of the sporopollenin following KEDVES (1989b), Fig. 1, p. 63.

Materials and Methods

The investigation material was collected by Dr. M. KEDVES on the 08.09.1998. in Szeged on a weedy piece of ground. Several partial dissolution and degradation experiments were carried out on these pollen grains. One of them, No: 1/7-1394 (10 mg pollen grain + 1 ml 2-aminoethanol during 24h + 10 ml 0.1% $KMnO_4$ during 24h on 30 °C) resulted a peculiar stage of the biopolymer structure of the sporoderm. Namely a number of the regular pentagon units of the quasi-crystalloid skeleton were degraded.

One of them was chosen for symmetry operations by the modified Markham rotation method (cf. KEDVES, 1989a). The results are presented in this contribution.

Results

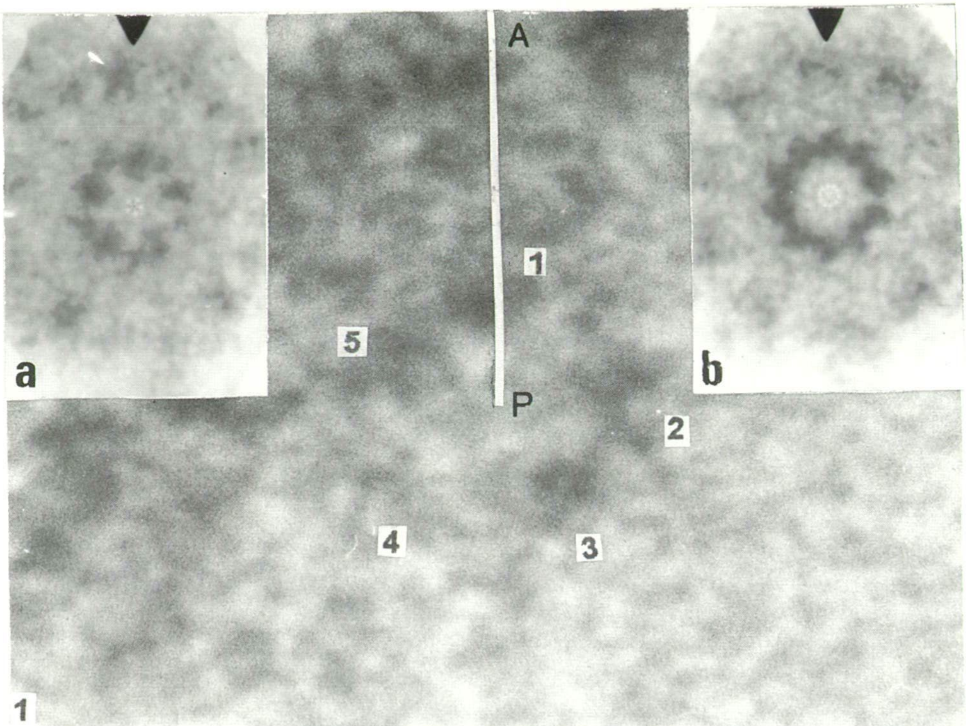
The transmission electronmicroscopical picture of the partially degraded ectexine of *Ambrosia artemisiifolia* (Plate 16.1., fig. 1) well illustrate that the biopolymer units in Angstrom dimension are not in a well preservation. On some parts in this picture molecular units sensu strictu are perceptible. A relatively very damaged regular pentagon was chosen for symmetry operations. The units of this regular pentagon are numerated, and the rotation axis PA is indicated. Picture 1a represents the result of the fivefold rotation (C.P.5.A.5.5.). The pentagonal rotation area is well shown its sides are parallel with those of the basic pentagon. But there are no characteristic positive or negative secondary points of symmetry around the rotation centrum. The result is similar at the tenfold rotation (Plate 16.1., fig. 1b). There are no characteristic points of symmetry around the rotation centrum except the first circle when there are ten dark points of symmetry. Around the rotation area there are ten clusters of cyclic molecules. One central molecule is surrounded by five other cyclic molecules, which are ressembling to another molecular Penrose unit.

In a highly magnified picture (Plate 16.1., fig. 2) all clusters of molecules are numerated altogether ten. Plate 16.2. illustrates in very highly magnified picture the results of the tenfold rotation. The biopolymer clusters 4 and 5 are well shown, and the surrounding cyclic molecules are numerated.

Discussion and Conclusions

As we have previously emphasized, the metastable quasi-crystalloid skeleton together with its stabilizing molecular systems are one component of the spore wall. These units may be building elements of the other kinds of highly organized elements such the helical structures in nanometer dimension (cf. ROWLEY et al. 1981).

In another paper we have demonstrated that the quasi-crystalloid structure may be present also on molecular level (KEDVES, TÓTH and FARKAS, 1993). But the regular pentagonal molecule was not in the rotation area of the biopolymer structure. Further molecular clusters were demonstrated from the partially degraded ectexine of *Encephalartos transvenosus* by KEDVES, PÁRDUTZ, TERBE and HORVÁTH (1999). The central cyclic molecule is surrounded by six cyclic molecules. The symmetry operation of the molecules of this cluster is in progress.



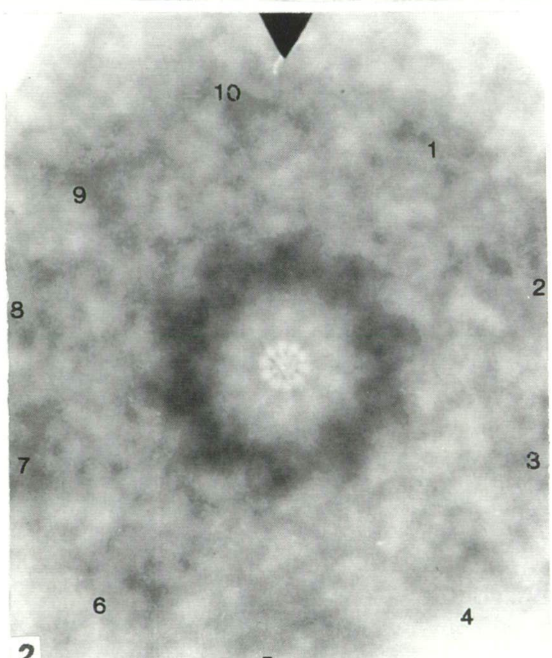


Plate 16.1.

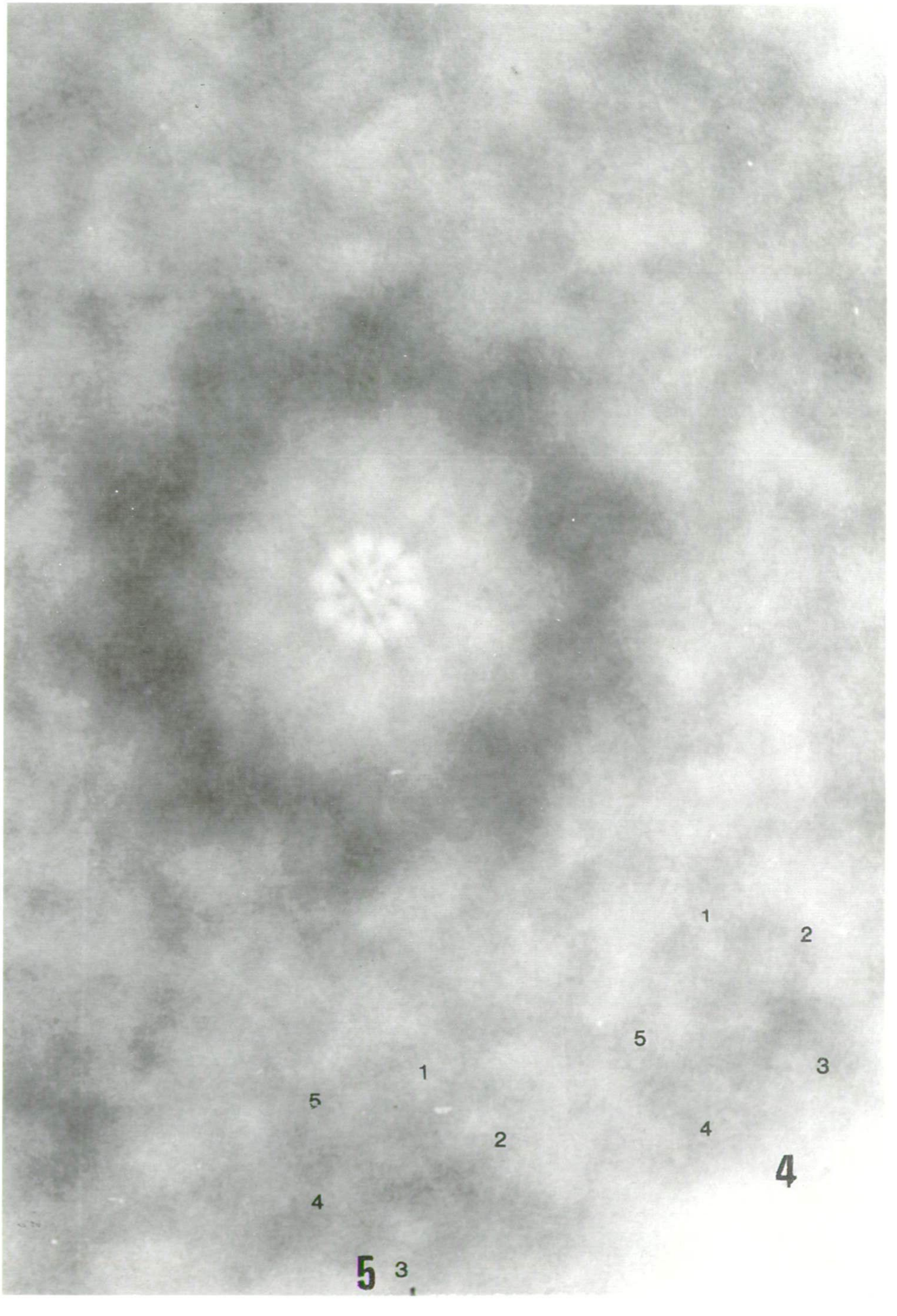


Plate 16.2.

Acknowledgements

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Plate 16.1.

1,2. *Ambrosia artemisiifolia* L., TEM picture of the partially degraded exine
1. Ultrastructure of the partially degraded ectexine. Experiment No: 1394, negative no: 7560, 2,500.000x.
1a,b. Rotation pictures 1,000.000x.
1a. C.P.5.A.5.5.
1b. C.P.5.A.5.10.
2. C.P.5.A.5.10, 2,000.000x.

Plate 16.2.

C.P.5.A.5.10 rotation picture 5,000.000x. The five surrounding units of the 4th and 5th Penrose-like molecular clusters are numerated.