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Shape and Surface Structure of the Magnetic Micro-Spherules from Permian and Triassic Bedded Chert

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(With 1 Table, 3 Figures and 5 Photoplates)

Abstract

A total of 801 magnetic micro-particles (including 792 magnetic spherules, 18 drop-shaped magnetic particles and one rocket-shaped magnetic particle) were collected from Permian and Triassic bedded chert of the Tamba-Mino Belt, Southwest Japan. Their shapes and surface structures show the characteristics of rapid quenching.

The surface structures of the magnetic micro-spherules are classified into five types: A. Random mosaic, B. Dendritic mosaic, C. Feather crystal, D. Scaly, and E. Cracked. These types show gradational changes in sequences A-B-C, A-B-D-E, and C-D-E. The mixed occurrence of three types (A, B, C) on a rocket-shaped particles indicates their transitional interrelationships as well as the conditions of their formation. Type A is considered to be the highest temperature phase. The formation of the surface structures are considered to have been controlled or affected by a complex mixture of the following factors: the manifestation of magnetite crystal structure, mechanism of crystal nucleation, degree of supercooling, and oversaturation.

Key Words: Micro spherules, Cosmic dust, Surface structure of magnetic micro-spherules, Magnetite, Bedded chert

I. Introduction

Many magnetic micro-spherules have been discovered from the deep-sea sediments. They are considered to be cosmic spherules, and their concentrations, size distributions, chemical composition, cosmic-ray produced nuclides, and genesis have been studied (e.g. YAMAKOSHI, 1979, 1984; BROWNLOW *et al.*, 1966; PERKIN *et al.*, 1980). However, one of the fundamental characteristics of the micro-spherules, i.e., their surface structures, which can be observed only by taking their SEM photographs, has not been discussed in detail systematically. It is thought that the process of creation of the magnetic spherules can be studied through the analysis of their surface structures. The present study presents detailed descriptions and a classification of surface structures of 801 magnetic micro-particles collected from Permian and Triassic bedded chert of Southwest Japan and discusses first on the relationship among different surface micro structures and second, on the controling factors which resulted in different surface micro structures.

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II. Extraction of the Magnetic Spherules

A total of 801 magnetic spherules and semi-spherical particles was collected from ancient pelagic sediments, i.e., rock samples of Permian and Triassic bedded cherts in the Tamba-Mino Belt, Southwest Japan (Fig. 1) (IWAHASHI *et al.*, 1991). Some of the chert samples include radiolarian fossils which are useful in determining the geological age of the cherts. They range from early Middle Permian to Middle Triassic (Fig. 2). ISHIGA (1986), YAO (1982) and other related works were used to identify the radiolaria and to determine their geological age.

IWAHASHI *et al.* (1991) gave description and discussion of grain size distribution, chemical composition, and mass influx of the micro particles. The chemical composition of some of the micro particles, analyzed by EPMA on the cross section, indicates that they are mostly magnetites; they differ from terrestrial magnetites in the abundance of minor elements, but resemble some cosmic magnetites: especially do "magnetite spherules" (EL GORESY, 1976, TOMEOKA *et al.*, 1989) in carbonaceous chondrite. According to IWAHASHI *et al.* (1991), the characteristics of grain shape, size distribution, surface structure and chemical composition of the magnetic micro-particles were considered in total to indicate that all of them are the cosmic spherules.

The collection of magnetic micro particle was carried out as follows.

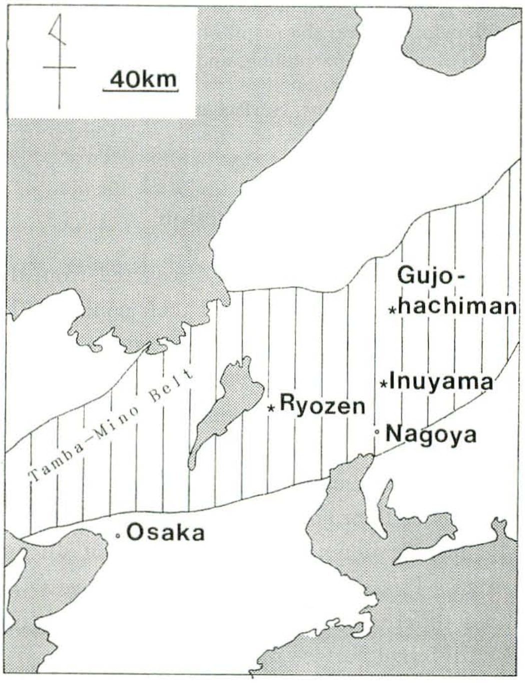

Fig. 1 Map of the area sampled.
Stars indicate the locations of sampling.

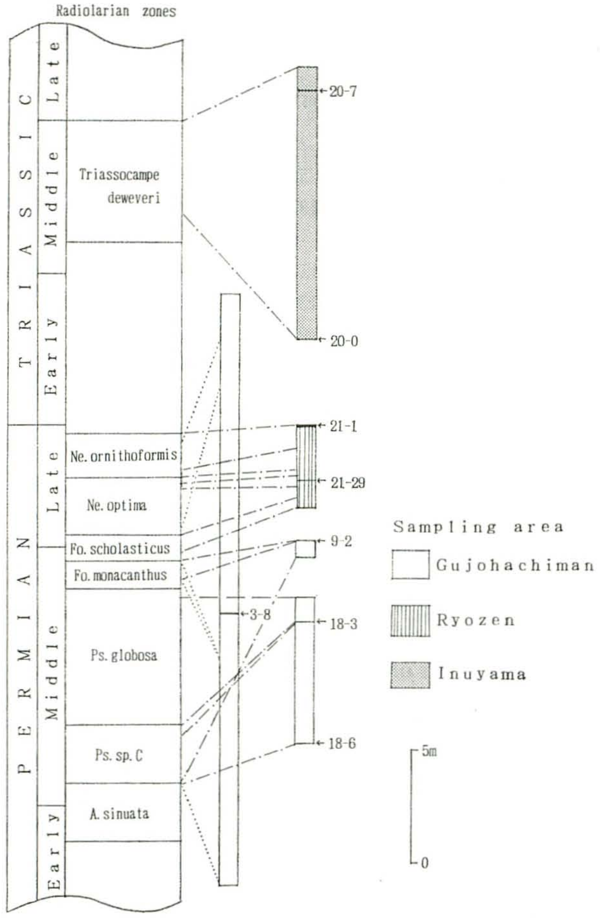

Fig. 2 Columnar sections of chert specimens which were used for this study and their possible limit of geological age based on radiolarian fossils. Radiolarian zones are from Ishiga (1986) and Yao (1982).

i) Put a chert piece between copper plates and crush into powder by a vice.
ii) Pass the chert powder through a sieve (35 mesh, opening 420 microns). Leave out bigger fragments.
iii) Take an accurate measurement of the chert powder.
iv) Pick up the magnetic particles from the chert powder using a magnet.
v) Separate heavy particles from the remaining chert powder in tetra-bromethane for 30 minutes.
vi) Mix the heavy particles with magnetic particles, and clean with acetone or alcohol in a supersonic washer.
vii) Collect dark spherical and semi-spherical micro-particles under a binocular microscope $\times 80$).

Table 1 Number of magnetic micro-particles collected from chert specimens.

chert No.	sampling area	numbers(N)		
		spherule	drop-shape	rocket-shape
20–7	Inuyama	27	0	0
20–0	Inuyama	59	3	1
21–12	Ryozen	137	4	0
1–29	Ryozen	0	0	0
3–8	Gujohachiman	158	4	0
9–2	Gujohachiman	137	1	0
18–3	Gujohachiman	20	0	0
18–6	Gujohachiman	254	6	0
total		792	18	1

Magnetic micro-spherules and semi-spherical particles (drop-shaped particles and a rocket-shaped particle) were collected by this method (see Table 1). All the magnetic micro-particles were observed using a scanning electron microscope (SEM), and their photographs were taken.

III. Observation of Grain Shape and Surface Stryctures

1. Grain Shape

The grain shapes of all the collected magnetic micro-particles are grouped into two groups: spherical particles (spherule) and semi-spherical particles (drop-shaped and rocket-shaped) (Plates 1–4). Most of them are spherules (see Table 1). Almost half (1/3–1/2) of the spherules, some of the drop-shaped particles and the lone rocket-shaped particles are hollw. Fragments of hollw spherules were also discovered (Plate 4), and the insides of such hollow spherules can be observed. They show baby dendrites growing from the exterior to the interior of the spherules.

2. Surface Structures

(a) Spherules

A SEM image magnifies the various patterns seen on the surface of the magnetic spherules: the winkles like the surface of a brain, branches or feathers, cross stripes, and cracks. The surface structures of spherules are classified into five types: A, Random Mosaic; B, Dendritic Mosaic; C, Feather Crystal; D, Scaly; E, Cracked (Plates 1–3). Intermediate types between some of the above were also recognized. The spherules rarely carry several types in one particle. Among all particles collected, Type B and an intermediate A-B Type occupy more than 80 percent (some spherules have so many blots that their surface structures were impossible to be classified). Types D and E are rare. Type C spherules often carry spinel-twins. Type D spherules represent the accumulated structure of plate trigonal crystals. Intermediate types between A and E, B and E, and A and

C were not observed. The existence and non-existence of intermediate types is consiseteed to reflect the gradational changes between the five types (Plate 5).

(b) Drop-Shape

Surface structures on the drop-shaped particles belong mostly to Type D (Plate 2).

(c) Rocket-Shape

The only one rocket-shape magnetic particle (No. 20–0–1) shows a hollow structure and looks like a hollow spherule with a short skirt. This particle has three types of the surface structures (A, B and C) mixed together. Type A occurs on the head and projections on the body. On the main part of the body, Type B occurs on the front and Type C on the middle and back (Plate 4).

IV. Discussion

1. Grain Shape

(a) Spherule

Generally, liquid congeals by rapid quenching in air or water and forms spheres, because liqupies the smallest surface area in the sphere-form, and preserves it by rapid quenching. The atmization process applies this theory and makes metal micro–shperical particles by blowing a noble gas jet on to the metal liquid and quenches it (SINBA and MITANI, 1978). On the other hand, heating a material to steam in an arc furnace (about 1000°K) and quenching it in noble gas, congelation and fusion make sub-micro spherules (HAYASHI *et al.*, 1988). However, this process, when carried out under gravity-free conditions, makes bigger spherules; and if the quenching is in a gas which includes oxygen, the steam congeals as oxide spherules. For example, iron as hypermagnetite (HAYASHI *et al.*, 1988). This process reminds magnetite spherules in carbonaceous chondrite, which are considered to have congealed from primitive gas in the solar system; it may have the relevance to the creation of the magnetic micro-spherical particles.

In the case of the artificial sub-micro spherules, the steam concentrates to a liquid sphere and congeals from one nucleu on the exterior; the crystal spreads over the surface of the sphere, with crystallization to the interior. The exterior crust is bigger than the genetic non-congealed sphere. The fragments of hollow spherules show dendrites growing to the interior, and this indicates that the hollow spaces in the spherules existed from their birth. The fact that artifical, atomized iron powder is ofter hollow also supports the above indication.

(b) Drop-Shape

SASAKI (1983) also discovered drop-shaped iron particles in deep-sea sediments and noted that many drop-shaped iron particles were discovered in the crater of the Sikhote-Alin iron meteorite (KRINOV, 1964). He indicated that the drop-shaped particles had been created from falling iron meteorites.

Drop-shaped micro particles look just like drops. Liquid forms a drop-shape for a moment when torn from a mass of melting material. If rapid quenching takes place at that time, congelation maintains the drop-shape. Considering surface structures, drop-shaped particles may have experienced lower temperatures than many spherules, because they could not form spherical shape—the higher temperature condition may form spheres.

(c) Rocket-Shape

Rocket-shaped particles resemble hollow spherules in cross-section. This type of particle is important because the rocket-shape may indicate the falling position, with the spherical head to the ground, like a rocket or a shell falling down. Hydrodynamically, this posture causes the least frictional resistance. The rocket-shaped particles resemble the "flusk-shape" of KRINOV (1964).

2. Surface Structures

Various types of surface structures or intergrowth texture of magnetic micro-spherules have been reported from samples collected mostly from deep-sea sediments (e.g., PERKIN *et al.*, 1980; SASAKI, 1983; BORNHOLD and BONALDI, 1979; GAO *et al.*, 1987). But inter-relationships of the surface structures have been little discussed. Possible controlling factors for the formation of various types of surface structurestare discussed below.

(a) Difference in temperature conditions: the A-B-C sequence, Type D.

The rocket-shaped particle No. 20–0–1 carries three types of surface structure: A, B and C. The rocket-shaped particle is considered to have fallen with its spherical head to the ground. Thus, the head and side projection of the particle was its hottest part, and the back its coolest. The distribution pattern of the three types of surface structure on the rocket-shaped particle may indicate that Type A was formed under the highest temperature phase, Type B under a lower phase, and Type C under the lowest. Type D may also indicate a low-temperature phase, because drop-shaped particles carry Type D.

(b) Reflection of the crystal structure of magnetite.

Some spherules show the magnetite-crystal structure, for example, spinel-twins. Spinel-twins are obvious in Type C, especially in No. 21–1–105 (Plate 2) with (111) as the composition plane.

The surface structure of Type D appears to be the parallel group of octahedral crystals. Octahedral crystals are penetration twins of two tetrahedra on the (100) plane, according to the Mohs-Rose law (ORLOV, 1977). Magnetite crystals often occur as octahedra (ROBERTS *et al.*, 1974). Therefore, Type D is considered also to show the crystal structure of magnetite.

Dendritic crystals indicate very rapid cooling, often rapid congealing from gas. Types B and C may, therefore, indicate very rapid cooling, possibly from a gas.

(c) Crystal Nucleation

Type A and E appear to carry similar crystallites, but the crystallites in Type A are much smaller than 1 micron in diameter, whereas those in Type E are above 1 micron. Many occurrences of uniform nucleation denote a large degree of supercooling or overss-turation resulting in the formation of fine clystals. Type A is considered to be a high-temperature phase and, therefore, it might have been supercooled and oversaturated. On the other hand, if melting material was preserved in a constant state just below the melting

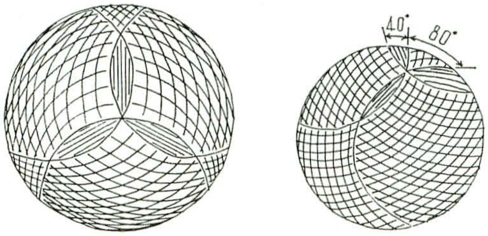

Fig. 3 A characteristic geometrical pattern obsereved on some Type B spherules.

point, no crystal nucleation takes place and instead, crystals grow by spiral growth. The intermediate type D-E (No. 9–2-1) carries the spiral-growth structure in some of the crystallites (Plate 5.) Type E has bigger crystals than the other types. Therefore, it might have experienced a smaller degree of supercooling and oversaturation, resulting in some amount of nucleation.

There are some spherules, especially in Type D and sometimes in Type B, that reflect the enlargement of crystals from one point. That point may be the first nucleus. This kind of Type B, dendritically spread crystals display very beautiful geometrical patterns (Fig. 3). Good examples are 7 (No. 20–0–21) and 8 (No. 18–6–231) of Plate 1.

V. Conclusion

The shapes of the magnetic micro-particles—spherule, drop-shape, rocket-shape—indicate that these particles formed by rapid quenching from either liquid or gas, by ablation from melting material, and flight in the air.

Five types of surface structures show the gradational sequences A-B-C, A-B-D-E, and C-D-E. Their surface structures are thought to reflect the following controlling factors affected during growth: the manifestation of magnetite crystal structure, the mechanism of srystal nucleation, the degree of supercooling cooling and oversaturation, and the temperature condition. From the rocket-shaped pariticles, it is clear that Type A is the highest temperature phase and Type C is the lowest in the A-B-C sequence. Type A carries very fine crystallites (sub-micron), indicating a large degree of supercooling and oversatuation. Type C often carries spinel-twins. Surface structures of Type D are thought represent the parallel group of octahedral crystals, which are common for magnetites, and may indicate a low-temperature phase. Type D usually shows enlargement of a crystal from point, such a point indicating the first nucleus. Type E carries large crystallites. A spherule of the intermediatettype D-E shows spiral growth on a cristallite. This may indicate a small degree of supercooling and oversatuation.

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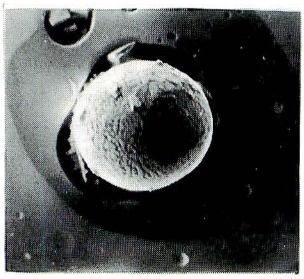
Plates 1～5

Explanation of Plate 1

Plate 1 Surface structures of spherules.

A	1:	No. 3–8–14,	d=16 microns
	2:	18–3–4,	23.3
	3:	18–6–191,	33.4
A-B	4:	18–6–193,	26
	5:	18–6–29,	20
	6:	9–2–150,	10.8
B	7:	20–0–21,	16.7
	8:	18–6–231,	23.3
	9:	18–6–244,	25.3
B-C	10:	3–8–118,	16
	11:	18–6–204,	25.3
B-C-D	12:	18–6–49,	41

A



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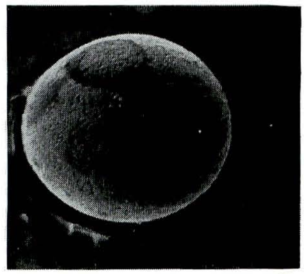

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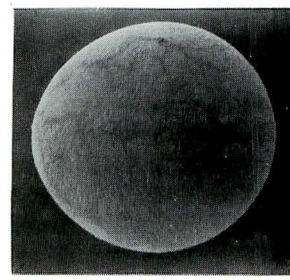

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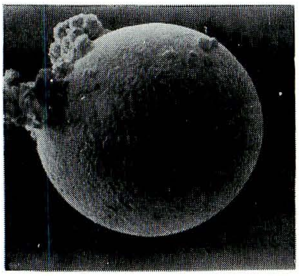
A-B



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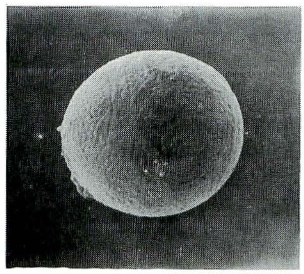

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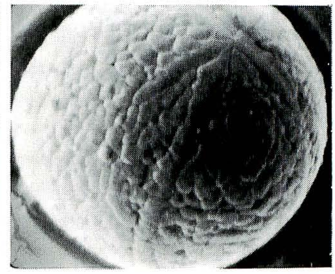

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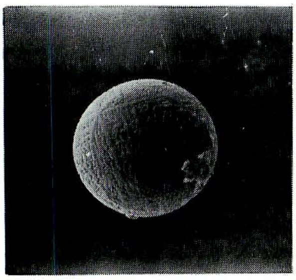
B



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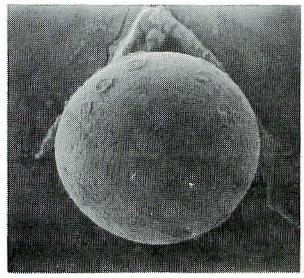

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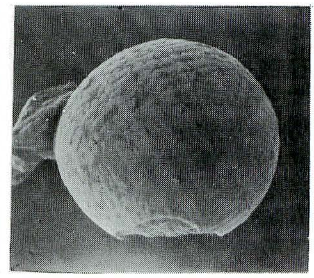

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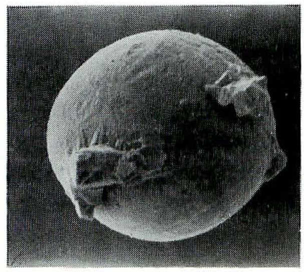
B-C

10



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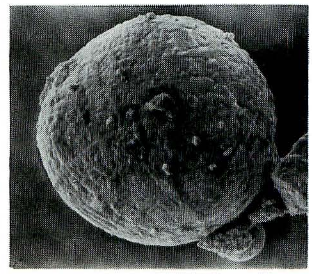
B-C-D



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Explanation of Plate 2

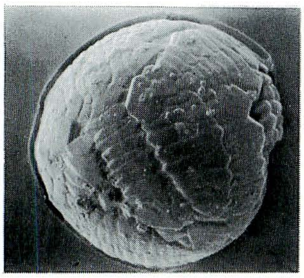
Plate 2 Surface structures of spherules and drop-shapes.

C	13:	No.	21–1–105,	d=35.6 microns
	14:		3–8–131,	34
	15:		18–6–216,	17.2
C-D	16:		18–6–44,	24.7
	17:		18–6–260,	24
D-E	18:		20–0–15,	24
D	19:		21–1–54,	30
	20:		20–0–17,	26.7
E	21:		20–0–3,	24

Drop-shaped particles (width of the photo: 34.7 microns).

22:	No.	18–6–99,	type D
23:		21–1–126,	C-D
24:		18–6–149,	B-C-D

C



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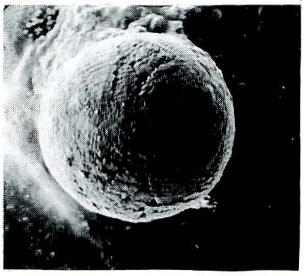

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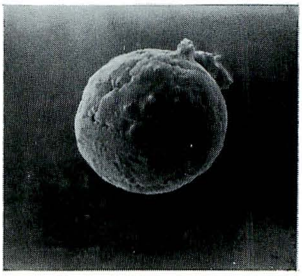

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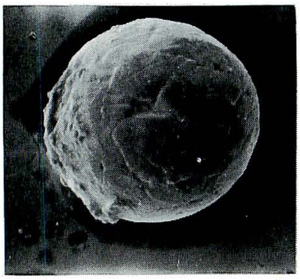
C-D



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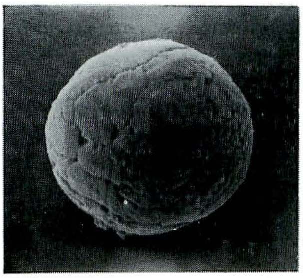

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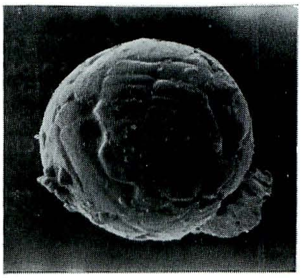
D-E



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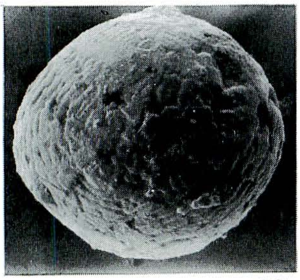
D



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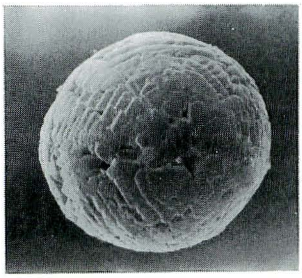

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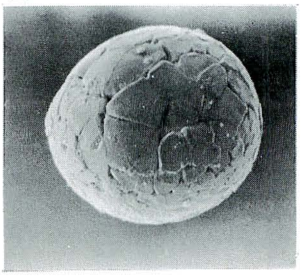
E



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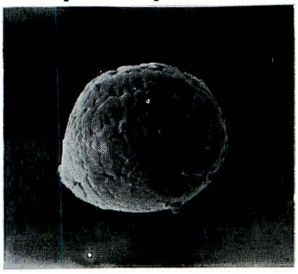
Drop-shape



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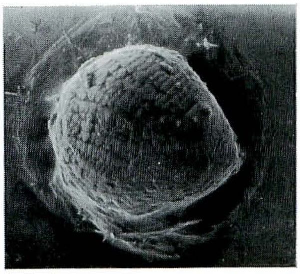

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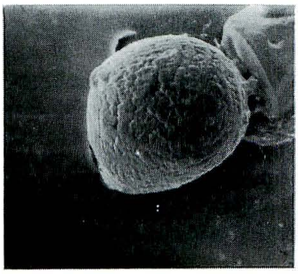

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Explanation of Plate 3

Plate 3 Enlarged view of some surface structures (width of the photo: 12.8 microns).

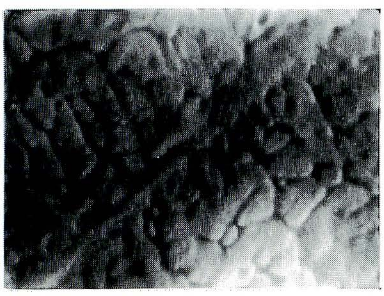
Type A (Random mosaic)
1: No. 20–0–23
2: 20–7–9
Type B (Dendritic mosaic)
3: No. 20–0–1
4: 20–0–8
Type C (Feather crystal)
5: No. 20–0–1
Type C-D
6: No. 20–0–80
Type D (Scaly)
7: No. 20–0–17

A

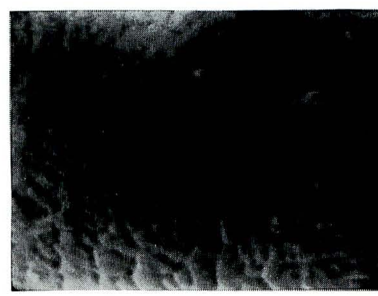
1

2

B

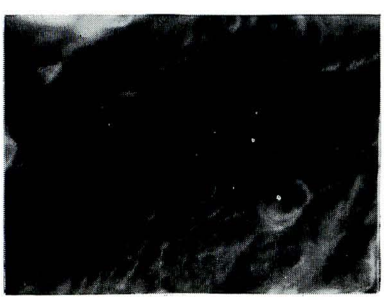


3



4

C

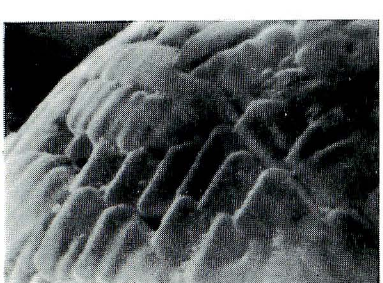


5

C·D

6

D

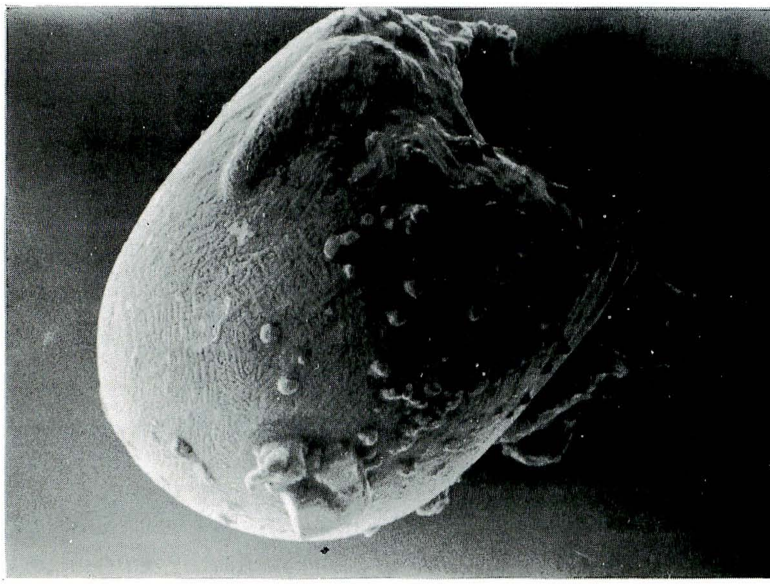


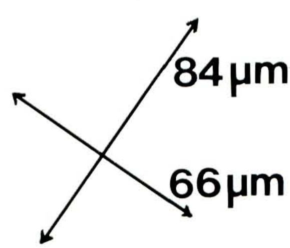
7

Explanation of Plate 4

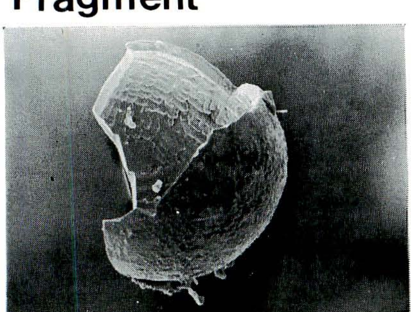
Plate 4 Rocket-shaped particle (No. 20–0–1) and some of the fragments of the hollow spherules (width of the photo: 84 microns).

Rocket-shape (No.20-0-1)



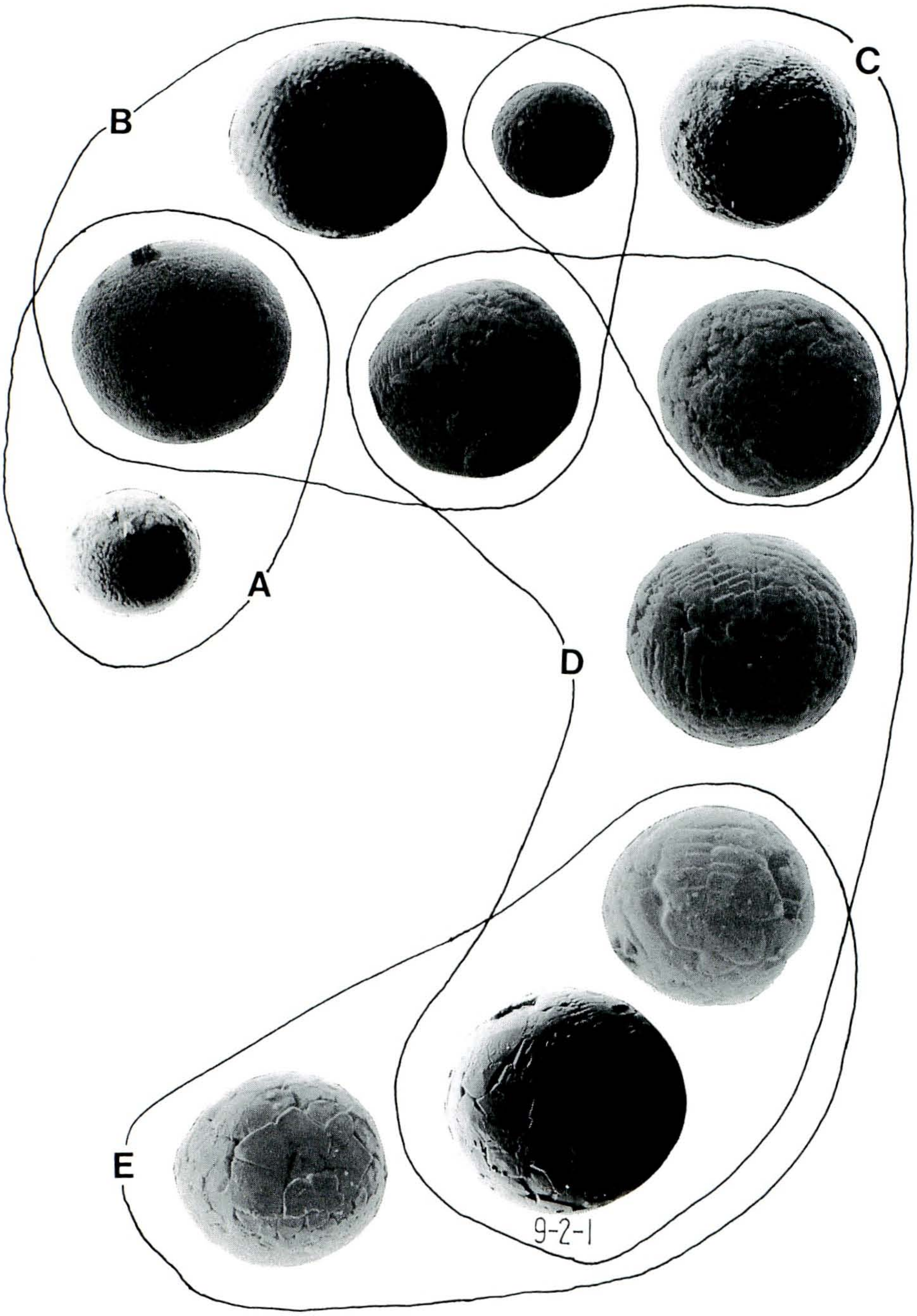


Fragment



Explanation of Plate 5

Plate 5 Gradational changes among five types: existence and non-existence of the intermediate types.


A
B
C
D
E
9-2-1