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MAGNETIC MICROSPHERULES IN PERMIAN AND TRIASSIC BEDDED CHERT FROM SOUTHWEST JAPAN

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Abstract: Magnetic microspherules collected from the Permian and Triassic bedded cherts in Southwest Japan were studied. The size range, as estimated from 801 microspherules, vary from 3 to 100 μm, with a peak size distribution between 10–20 μm. Three broad shapes are recognized: spherical-, drop- and rocket-shaped, all categories including hollow particles. The surface textures as studied from scanning electron microscope show five dominant varieties: random mosaic, dendritic mosaic, feather crystal, scaly and cracked, with intermediate textures between these types. Chemical analyses of representative microspherules by electron microprobe indicate that they are mostly composed of magnetite, resembling in composition with microspherules of cosmic origin from other regions and magnetite grains in carbonaceous chondrites. Consideration of the origin of the observed morphologic, textural and chemical parameters in these microspherules, along with the available data on experimental synthesis of ultramicro iron particles, preclude an origin by volcanogenic processes and strongly suggest a cosmic origin. Knowledge of the rate of chert sedimentation allows us to make an empirical calculation on the rate of microspherule sedimentation. We compute that the fall of microspherules on the earth's surface occurred at the rate of about 1 t/day during Permian, while it increased to about 3 t/day during the closing of the era and the Triassic.

1. Introduction

Unusually high concentrations of fine-grained extraterrestrial particles deposited on the earth's surface have been noted in a variety of environments. Opinions are divided on the origin of these cosmic dust, although many workers link them with dust from comets, or ablation debris from meteorites (BLANCHARD and DAVIS, 1978; YAMAKOSHI, 1979). Though black, shiny sub-millimeter sized spherules were discovered as early as 19th century, most of the inferences on their origin remained theoretical extrapolations mainly due to the lack of analytical advances in determining the compositional characteristics of such microscopic particles. Several studies have recently concentrated on the cosmic spherules from air, continental ice sheets and deep-sea sediments (BORNHOLD and BONARDI, 1979; ROBIN *et al.*, 1988). However, fundamental to the understanding of cosmic dust and their possible relationship with earth's evolutionary history, including periodic mass extinctions of livings, is the knowledge of the deposition of microspherules in the geologic past. Such information could be extracted from sedimentary rocks that were deposited at varied time intervals during the geologic history, not so much attention having been paid to this aspect (CROZIER, 1960; MUTCH, 1966; GAO *et al.*, 1987).

In Japan, the most potential litho-unit for such studies are bedded cherts which occur in various geologic horizons dating from Permian. The present study is aimed at documenting the data on magnetic microspherules recovered from Middle-Late Permian to Middle Triassic chert. This investigation has concentrated on deciphering the microspherule abundance, size distribution parameters, shape, surface structure, chemistry, and the amount of microspherules falling onto the earth.

2. Sampling

Following the geologic outline given in WAKITA and OKAMURA (1982), WAKITA (1983), ISHIGA *et al.* (1982) and YAO *et al.* (1980), bedded chert samples were collected from three main areas, namely, Gujohachiman, Ryozen, and Inuyama (Fig. 1). In all these areas, the chert units are included as olistostromes in younger Mesozoic rocks. A total of 121 specimens of bedded cherts were collected and examined, some of them containing radiolarian fossils useful for determining geologic ages. The age of the samples used for this study ranges from Permian to Triassic based on their radiolarian record (ISHIGA *et al.*, 1982; YAO *et al.*, 1980 and other related works).

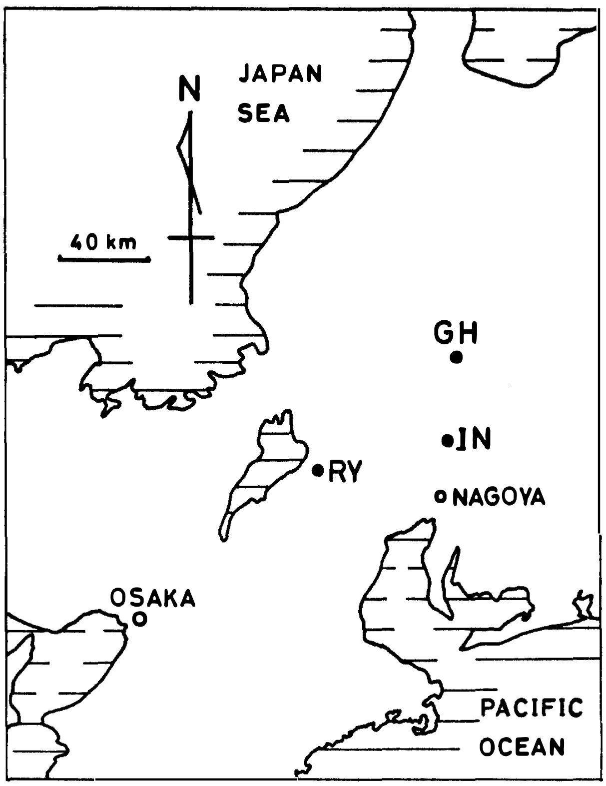

Fig. 1. Sampling localities of the Triassic bedded chert. GH: Gujohachiman; RY: Ryozen; IN: Inuyama.

Out of these, eight specimens were selected for detailed study of magnetic microspherules. They contain radioralian fossils and therefore are appropriate to obtain the correct age of deposition. Similar to meteorite classification into iron, stony and stone-iron, it is considered that stony or glassy categories of cosmic spherules exist, in addition to their more common occurrence as 'black magnetic' spherules (YAMAKOSHI, 1979). The present work, however, concentrates only on the magnetic spherules, not only because they are candidates for easier separation but also due to their prime importance in a fundamental study of this kind.

3. Methodology

The rock fragments of about 200 g each were carefully collected from each bedded chert layer of about 10 cm thick, in such a way that each sample represents the complete thickness of individual layers. The spherules were then separated from these rock specimens. First, the rock sample was put in between two copper plates and was crushed using vise. The crushed powder was then sieved (35 mesh, sieve opening 420 μm), leaving out the bigger fragments. The weight of the $<420\ \mu$m fraction was recorded, which range from about 9 to 15 g. Using a hand magnet, magnetic particles were then carefully separated from the sieved fractions. The heavies were separated from the remaining powder using techniques of heavy-liquid separation (tetrabromethane for 30 min). Through the latter procedure, magnetic microspherules which were not retreived by hand-magnet were able to be separated along with other heavies. These heavies were then mixed with those separated by hand-magnet and then cleaned with acetone or alcohol in an ultrasonic washer, in order to separate the adhering particles for easy hand-picking. The cleaned magnetic particles and heavy mineral fraction were then examined under a binocular microscope (spherules being visible normally under the magnification of 80 times) and all the visible spherules were then carefully separated by hand-picking using a moist slender bristle. The minimum measurable size of microspherules in the present study is 3 μm, and it is believed that there is almost no microspherules having the diameter over 5 μm left uncollected. Out of the many microspherules recovered, only a few (approximately 3%) were found to be fragmented. The fragments were part of the shell of the hollow microspherules. About half of the microspherules of the present study are considered to be hollow particles, judging from the characteristics of the fragmental particles as mentioned above and the cross sections of complete microspherules obtained during the preparation of the chemical analyses as mentioned below.

In order to study the morphological parameters including shapes and surface structures of the spherules, they were mounted on aluminium stubs and examined using a scanning electron microscope. Selected representative spherules were then dismounted, moistened by xylene and mounted over slide-glass using Petropoxi 154 glue. They were then polished using fine carborundum and diamond paste, although a high degree of brilliance could not be achieved by polishing due to the inherent limitations of sample preparation when dealing with micron-sized particles. The resultant cross-sections of the spherules were first examined in detail under the microscope and then subjected to electron probe microanalysis (EPMA) at the National

Institute of Polar Research, Tokyo (JEOL Superprobe).

4. Results

A total of 801 microspherules were collected from the eight chert specimens. The details of sampling sites, age, weight of powdered cherts, and number of microspherules recovered are presented in Table 1. The volumes of microspherules were calculated from their diameters (as measured from SEM photographs).

As to the fragments of microspherules, only those in which more than half of the complete particle were preserved have been taken into account to obtain the total volume. For volume calculation, the hollow spherules were assumed to have been filled.

4.1. Size distribution

The diameter of spherules ranges from about 3 to 100 μm (Fig. 2). The peak size distribution lies in the range of 10–20 μm. This size range is consistent with those reported from Mesozoic–Paleozoic sediments by CROZIER (1960), although they are considerablly smaller than those from recent sediments. However, the distribution pattern showing the decrease in frequency towards smaller size is remarkably different from that of CROZIER (1960) and is also not consistent with the general understanding that the frequency may increase towards smaller size (*e.g.*, YAMAKOSHI, 1979). Those over 20 μm in diameter decrease logarithmically. We do not consider that the decrease in numbers less than 10 μm is due to sample bias, arising from difficulty in recovering smaller particles. In the chert specimens which contain over 100 spherules (except sample 18–6, which is mixed with dolomite), the $\log_{10}$F (size frequency) is proportionate to $-2.8 \log_{10}$D (diameter) above 10–20 μm (Fig. 3). The peak positions of the spherule diameter differ slightly with respect to individual rock specimens. Excepting one specimen, which is a dolomite-mixtured chert is a broad positive relationship between the thickness of the chert layer and the peak diameter of microspherules, although their relationship is not well defined.

4.2. Shapes and surface structures of the microspherules

The shapes of all the collected particles can be broadly grouped into spherical, drop-shaped, or rocket-shaped particles (Fig. 4). Majority of them corresponds to the category of spherical particles (*cf.* Table 1). We could observed the internal textures of spherical particles in some cases by studying the inside portions of fragmented hollow particles (Plate 1). SEM studies have brought out in detail the surface characters of microspherules, which show wrinkles like the surface of a brain, branches or feathers, cross-stripes and cracks. The observed surface structures can be classified into five dominant types: (A) random mosaic, (B) dendritic mosaic, (C) feather crystal, (D) scaly and (E) cracked. Intermediate categories were observed between some of the above types. Howerver, no microspherules showing intermediate surface characters between A and C, A and E, C and E and B and E were observed (Plate 2). Among all the particles collected, those having a dendritic mosaic texture (type B) and intermediate between random and dendritic type mosaics comprise more than

Table 1. Collection of magnetic microspherules from 8 chert samples.

Sample No.	Sampling area	Age	Thickness of chert layer (cm)	Weight of sieved fraction of chert (g)	Numbers (N)			Spherule contents	
					Spherical shape	Drop-shape	Rocket-shape	(N/g)	($\times 10^{-8}$ cm^3/g)
20–7	Inuyama	Middle Triassic	6.5	9.3446	27	0	0	2.89	6.208
20–0	Inuyama	early-Middle Triassic	8.5	9.1183	59	3	1	6.47	4.833
21–1	Ryozen	late-Late Permian	12.5	10.5300	137	4	0	13.01	20.574
21–29	Ryozen	middle-Late	5.0	15.1165	0	0	0	0	0
3–8	Gujohachiman	early-Late Permian	8.0	15.5225	158	4	0	10.18	7.490
9–2	Gujohachiman	late-Middle Permian	4.0	15.6837	137	1	0	8.74	5.995
18–3	Gujohachiman	middle-Middle Permian	3.5	10.2537	20	0	0	1.95	0.657
18–6	Gujohachiman	early-Middle Permian	1.3	13.4555	254	6	0	18.83	8.819

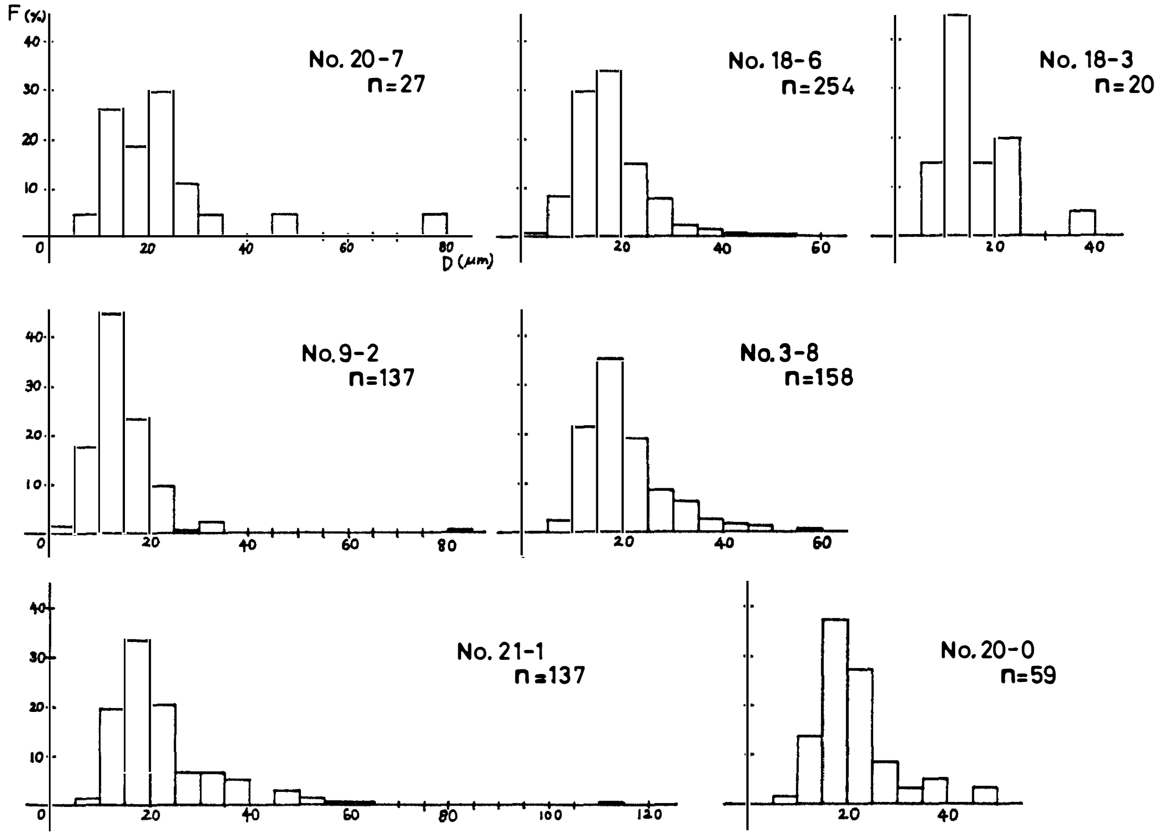

Fig. 2. Size distributions of the magnetic microspherules from Permian and Triassic bedded cherts.

80%. Types D and E are relatively rare.

Almost all the microspherules are composed mostly of magnetite as mentioned below (see section on spherule chemistry). Some of them show magnetite crystal structure with octahedral crystal habit as also observed by other workers (ROBERTS *et al.*, 1974). Spinel twins are common among type C with the (111) compositonal plane. Types B and C are considered to be dendritic crystals. The surface structure of type D particles resembles parallel groups of octahedral crystals. Types A and E appear to contain similar crystallites, although those in type A are much smaller (<1 μm) than in E (>1 μm). One specimen of the intermediate type between D and E has spiral growth structure on a part of the crystallite (No. 11, Plate 2). The incipient growth stages are spectacularly preserved in some of the type B spherules. The crystals spread dendritically in a distinct geometric pattern (No. 4, Plate 2).

The surface structure of the drop-shaped particles mostly belong to D-type (Nos. 3 and 4, Plate 1). A lone rocket-shaped magnetic particle (No. 5, Plate 1) appears like a hollow spherule with a short skirt. This particle shows a combination of the three types of surface structures A, B and C. At the pointed "head" of the particle, as well as on the sharp projections on the "body", type A structure predominates. Type B is seen on the other smoother portions, towards the head-ward side and type C towards middle and back.

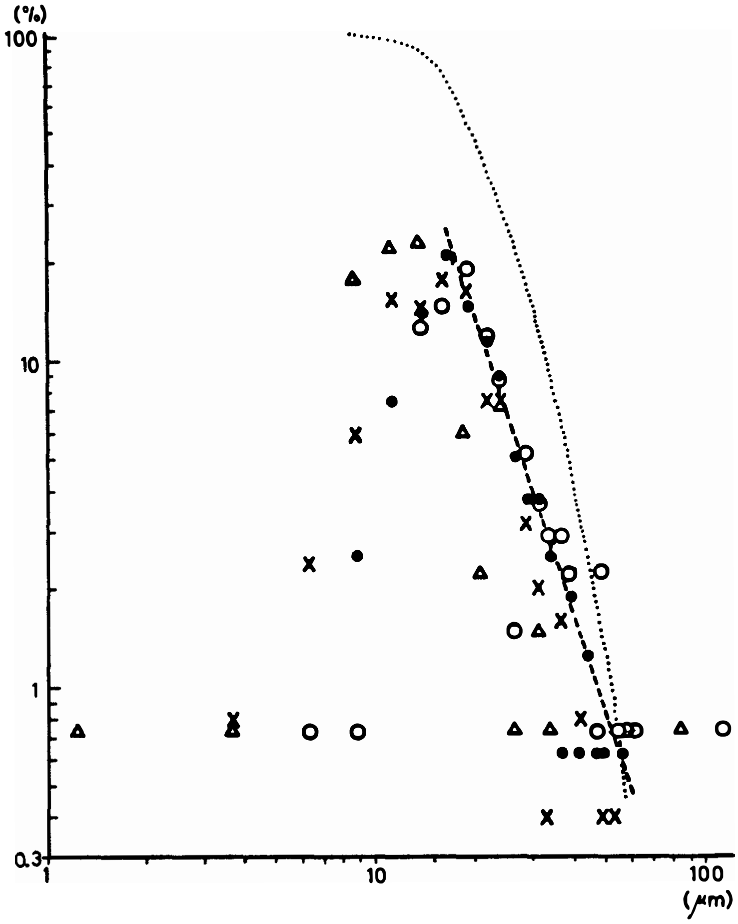

Fig. 3. *Logarithmic plots of size distributions of magnetic microspherules from some bedded cherts.*
Cross: 18–6; Triangle: 9–2; Open circle: 21–1; Solid circle: 3–8. Size frequency is proportionate to −2.8 log diameter, as shown by straight chain for sample 3–8. Dotted line shows cumulative curve for the same sample.

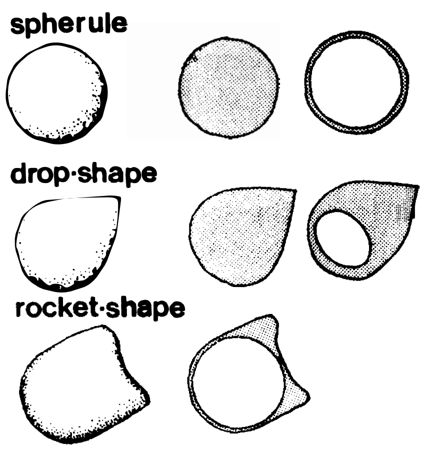

Fig. 4. *Shapes (left) and cross-section (right) of magnetic microspherules.*

5. Spherule Chemistry

Thirty-nine representative microspherules were analyzed on over 50 points including the central portion selected as the center of cross section of the particle, shell of the particle detected from its cross section, and surface of particles. The low probe totals are attributed to the analytical conditions of less surface brilliance of the spherules as mentioned above. The total oxide value of the analyses mostly range from 33 to 91%, with only 20 showing total oxides of over 80%. When a part of FeO is recalculated into Fe_2O_3 in these analyses, those with total oxides of over 90% becomes nearly 100%. The low totals in the iron oxide values mostly are due to the analytical conditions and need not necessarily indicate the existence of undetected elements. Table 2 gives 17 analyses and average of the central portions of microspherules whose total oxide value is over 80%. These compositions translate mostly into magnetite, when given allowance for the analytical constraints as mentioned above. Characteristic surface structures of some of the microspherules are consistent with this identification. However, we do not eliminate the possibility of existence of less oxidized or more oxidized phases admixed with magnetite.

Table 3 gives the analytical result of surface and cross section of shell of microspherules. Although the analytical result may not be of high quality as indicated by the total oxide values less than 86%, they provide indication of compositional heterogeneity among a microspherule. In comparison to the average chemical composition of the central portions, it is noted that all of them are slightly depleted in Fe, and that many of them are enriched in other elements. Depletion in Fe may reflect the increasing oxidation state in some particles and cation substitution of Fe in others. Enrichment in other elements may be attributed either to the primary

Table 2. Chemical composition of the central portion of cross sections of magnetic microspherules.

Specimen No.	SiO_2	TiO_2	Al_2O_3	Cr_2O_3	FeO	MnO	NiO	MgO	CaO	Na_2O	K_2O	P_2O_5	Total
18–6–6	0.24	0.00	0.09	0.17	88.52	0.63	0.00	0.00	0.08	0.01	0.00	0.00	89.74
18–6–17	0.68	0.00	0.11	0.01	87.42	0.38	0.02	0.00	0.07	0.00	0.01	0.01	88.71
18–6–30	0.22	0.00	0.04	0.21	79.74	0.59	0.03	0.01	0.08	0.00	0.02	0.00	80.94
18–6–49	1.39	0.00	0.22	0.04	84.31	0.35	0.00	0.11	0.07	0.09	0.00	0.01	86.59
18–6–58	0.26	0.02	0.02	0.23	87.20	1.04	0.07	0.02	0.08	0.00	0.06	0.15	89.15
20–0–8	0.15	0.00	0.46	0.14	87.04	0.47	0.07	0.05	0.07	0.04	0.03	0.07	88.95
20–0–11*	0.36	0.03	0.13	0.05	88.34	0.37	0.08	0.04	0.01	0.00	0.01	0.14	89.56
20–0–13	0.26	0.00	0.12	0.03	89.79	0.48	0.00	0.03	0.10	0.05	0.00	0.10	90.96
20–0–17	0.23	0.04	0.10	0.23	90.11	0.42	0.11	0.00	0.04	0.00	0.01	0.11	91.40
20–0–36	0.15	0.00	0.05	0.03	90.53	0.52	0.12	0.01	0.07	0.00	0.02	0.01	91.51
20–0–60	0.84	0.00	0.16	0.14	86.68	0.50	0.03	0.00	0.05	0.03	0.01	0.00	88.44
20–0–66	0.41	0.00	0.21	2.04	86.97	0.35	0.01	0.00	0.10	0.02	0.02	0.00	90.13
20–7–5	1.21	0.00	0.15	0.03	82.20	0.37	0.07	0.00	0.05	0.01	0.00	0.01	84.10
21–1–6	1.13	0.00	0.15	0.16	83.05	0.48	0.01	0.04	0.09	0.09	0.02	0.12	85.34
21–1–105	1.41	0.00	0.21	0.40	79.67	0.63	0.04	0.00	0.11	0.01	0.01	0.06	82.55
9–2–1*	0.10	0.00	0.32	0.20	88.59	0.61	0.00	0.01	0.00	0.00	0.02	—	89.85
18–3–7	0.27	0.02	0.29	0.15	86.71	0.26	0.20	0.00	0.08	0.01	0.02	—	88.01
Average	0.57	0.01	0.17	0.25	86.29	0.50	0.05	0.02	0.07	0.02	0.02	0.05	88.02

*: Hollow particles. —: Not measured

Table 3. Chemical composition of surface and shell of magnetic microspherules.

Sample No.	SiO_2	TiO_2	Al_2O_3	Cr_2O_3	FeO	MnO	NiO	MgO	CaO	Na_2O	K_2O	P_2O_5	Total
Surface													
9-2-12	0.81	0.00	0.27	3.47	66.08	0.41	0.24	0.03	0.25	0.45	0.05	—	72.05
9-2-14	0.44	0.00	0.07	0.17	77.75	0.52	0.03	0.07	0.36	0.49	0.22	—	80.12
9-2-20	0.67	0.00	0.31	0.27	79.75	1.60	0.05	0.17	1.77	1.05	0.07	—	85.71
9-2-51	0.14	0.00	0.07	1.02	82.84	0.31	0.10	0.06	0.15	0.36	0.01	—	85.06
Shell													
18-6-39	0.87	0.00	0.35	0.64	68.18	0.00	0.00	0.08	0.46	0.01	0.05	0.07	70.71
20-7-1	1.20	0.01	0.27	0.01	61.23	0.00	0.10	0.00	0.03	0.01	0.00	0.00	62.85
Average	0.69	0.00	0.22	0.93	72.64	0.47	0.09	0.07	0.50	0.40	0.07	0.01	76.09

Table 4. Chemical composition of magnetic microspherules of present study compared with those from other studies.

	SiO_2	TiO_2	Al_2O_3	Cr_2O_3	FeO	MnO	NiO	MgO	CaO	Na_2O	K_2O	P_2O_5	Total
A	0.57	0.01	0.17	0.25	86.29	0.50	0.05	0.02	0.07	0.02	0.02	0.05	88.02
B	0.73	0.36	0.20	0.01	91.10	0.03	0.00	0.07	0.00	—	—	—	92.50
C	2.36	—	—	0.51	91.06	0.13	0.19	—	—	—	—	—	94.25
D	7.14	—	0.85	0.29	89.39	—	—	—	0.92	0.44	0.14	—	99.17
E	0.71	0.05	0.14	0.02	82.33	0.08	0.05	0.20	0.11	0.20	0.02	—	83.91

—: Not measured.

A: Average of 17 magnetic microspherules of the present study. B: Average of 4 terrestrial magnetite, DEER *et al.* (1962). C: Average of 4 analyses of SASAKI (1983) including hollow- and drop-type microspherules and shell portions of Fe-Ni spherules. D: Average of 5 microspherules after GAO *et al.* (1987). E: Average of 7 analyses of magnetite in C2 chondrite Y-790003 (after the courtesy of H. KOJIMA, unpublished).

chemical characteristics, or to the secondary alteration or contamination. Increase in Cr_2O_3 and NiO, however, may be noticed as primary characteristics, because these elements are impossible to be enriched during a secondary process.

Table 4 gives the comparison of average chemical composition of microspherules of the present study with some other published chemical analyses. In comparison to terrestrial magnetites (DEER *et al.*, 1962), the microspherules are conspicuously more abundant in Cr_2O_3. Terrestrial magnetites hardly include chromium oxide, except titanomagnetites. Also, the microspherules are contrasted from terrestrial magnetites by their near-absence of TiO_2 and presence of NiO.

Chemical composition of microspherules from deep-sea sediments have been studied by various workers. Some of the larger microspherules (>100 μm in diameter) from deep-sea sediments have a metallic core of Fe-Ni (ROBIN *et al.*, 1988), although spherules in deep-sea sediments comprise a wide size range and the smaller ones have not been chemically characterized as yet. SASAKI (1983) carried out chemical analysis, using electron microprobe analyzer, of a variety of microspherules; hollow, filled, Fe-Ni core and drop-shaped particles along with artificial spherules synthesized from an iron meteorite. The average chemical composition of the spherules of present study is comparable to SASAKI's analyses of hollow spherule, drop-shaped particle and the shell of artificial Fe-Ni core spherule (*cf.*, Table 4). SASAKI mentioned that many drop-shaped particles were found around the crater of Sikhote-Alin iron meteorite and their compositions were ascribed to iron meteorites.

Recently, GAO *et al.* (1987) reported microspherules from rocks of the Permian-Triassic boundary of the Shangsi Section in China. The chemistry of these microspherules analyzed by using electron microprobe, closely resembles those reported by us (*cf.*, Table 4). However, SiO_2 (4.54–11.51%) and CaO (0.66–1.28%) reported by GAO *et al.* are slightly higher than the values for the microspherules of present study (average 0.57 and 0.07 respectively).

Among the apparent extraterrestrial magnetites, those which chemically resemble the spherules of present study are the ones derived from carbonaceous chondrites. Spherules in carbonaceous chondrites are micron-sized minute magnetite grains which have internal structures indicative of origin from condensing gases. These are considered to be some of the latest material which had directly condensed from the solar nebula (EL GORESY, 1976). We have compared the chemical composition of magnetite grains in C2 chondrite Yamato-790003 analyzed by KOJIMA (personal communication 1990) with the average composition of spherules of present study. They are similar in chemistry, though the microspherule of present study are more abundant in Cr_2O_3 and while MgO and to a small extent Na_2O, are less abundant. Further, the magnetite grains in the C2 chondrite tend to show a systematic increase in MgO with SiO_2, though the microspherules of present study do not show such a relationship.

6. Discussion

The most pertinent aspect of any study of microspherules is that whether they truly represent cosmic particles. Often the evidences are not totally unambiguous, especially in the case of spherules separated from sedimentary rocks like the bedded cherts in the present case. Nevertheless, it is also true that there are no factors which deny their possibility of being extraterrestrial particles.

One of the major aspects which indicate a cosmic affinity for these particles is that spherical magnetites cannot exist in natural terrestrial environments unless they were once melted and quenched. Though there are authigenic spherical goethites, clearly the microspherules reported in this study are not goethites. Also, our morphological and textural studies indicate that most particles exhibit dendritic growth patterns on their surface. The other extreme possibility is that they are industrial artifacts; this is not the case of the present study dealing with chert of geologically old ages. However, industrial processes and related experiments are possible guidelines to understand how similar microspherules are produced naturally. Among processes which can produce spherical artifacts during industrial metallic powder production are shotting and atomization in the liquid powderings and carbonyl process in the physical and chemical powderings (WATANABE, 1976; SINBA and MITANI, 1978). Especially in the shotting and atomization processes, drop-shaped particles could also be produced. Atomized iron powder is often hollow, with a diameter range of about 60 to 300 μm, having oxidized surfaces (WATANABE, 1976). Smaller oxidized spherical powder is produced by melting, evaporating and quenching in gas (including oxygen) (HAYASHI, 1988; NIPPON KAGAKU KAI, 1985; ICHINOSE, 1988).

In nature, such environments conductive for the generation of spherical particles operate only in two possible settings, namely (1) at the eruption sites of volcanoes and (2) by the entry of particles into the atmosphere from outer space. Clearly, these magnetic microspherules cannot be linked with volcanic eruption primarily because as yet there is neither report which indicates that such spherules are produced by volcanic eruption, nor any finding of particles largely in association with volcanic sediments. Also, since magnetic microspherules have been recovered from deep-sea non-volcanic sediments of about 200 Ma million years ago), they should have been found in large quantitites in the present day volcanic sediments if they were of volcanogenic in origin. All these factors prompt us to reasonably assume that textural and morphological characteristics preserved by the microspherules were inherited during the time of their entry to the atmosphere from outer space.

The lack of enrichment in nickel in the microspherules of the present study, one of the most important elements in iron meteorites, can be explained by their incandescence in the air in the earth's atmosphere which contains oxygen. It has been noted that the fusion crusts of iron meteorites have the composition of magnetite, sometimes 100 microns thick (MASON, 1962). Recent experimental studies confirm that Fe more easily concentrates in oxide form than does Ni, which is hardly concentrated in its oxidized state (HAYASHI, 1988).

Experimental studies also throw much light (HAYASHI, 1988) upon the mechanism of formation and inheritance of the observed chemical characteristics of the microspherules. In the production of iron ultra-micro particles, when iron gas is passed through an atmosphere with oxygen partial pressure much greater than 0.013 Torr, iron oxide (hypermagnetite) grains are formed. However, when the partial pressure of oxygen was less than 0.008 Torr, iron oxide grains formed with a metallic iron core. It was also found that if the whole grain is hypermagnetite, the sudden introduction of oxygen cannot make the oxidation from Fe^{2+} to Fe^{3+}. By contrast, if the grain is both metal iron and iron oxide, it will be completely oxidized by the sudden introduction of oxygen (HAYASHI, 1988). These results have a direct bearing on the formation of the inner composition and structure of magnetic microspherules. It is also noted that the size distribution of the microspherules of present study is quite similar to the experimentally obtained distribution curve of metalic ultra-microspherules through vaporizing technique (ICHINOSE, 1988). Future experimental investigations on the synthesis of artificial microspherical and ultra-microspherical particles might further resolve the mechanism of formation and origin of natural spherules.

While the aforesaid arguments lead us to conclude that the magnetic microspherules from the bedded cherts of Japan represent cosmic dust, and were generated by heating and rapid quenching, the data also permit us to postulate the temperature history of these particles. We believe that the answer for this important question may be sought from the variations in the surface structures exhibited by these particles, especially from types A to B and to C as described by us. The rocket-shaped particle (No. 5, Plate 1), which resemble the "flask-shape" described by YAMAKOSHI (1984) is considered significant because it might indicate the orientation of the particle as it fell through the atmosphere, that is, with the spherical head towards the ground like in the case of a rocket or a shell falling on the earth. Hydrodynamic considerations indicate that such an orientation causes the least frictional resistance and so the head and the projection of the above-mentioned spherule became the hottest during the fall, while the tail portion remained cooler. This single particle has been observed to preserve three types of surface textures (types A, B and C), in direct correlation with the temperature variation between its front and rear portions. Thus type A structure formed under the highest temperature phase, type B in the intermediate zone and type C at the lowest temperature zone.

7. Spherule Sedimentation

If the rate of chert sedimentation is known, weight (metric tons) of the magnetic spherules accumulated per day on the earth (M) can be calculated from a simple

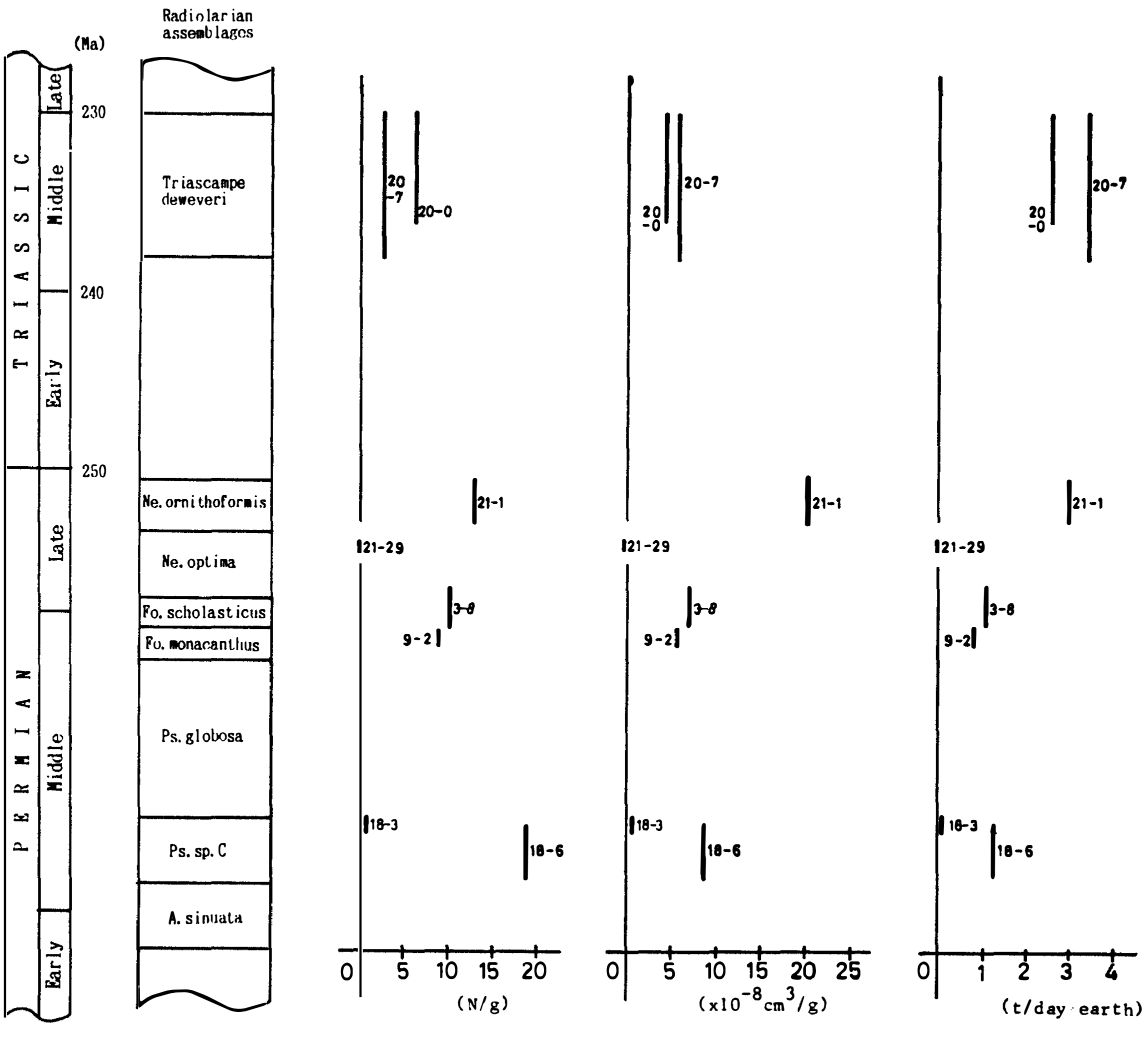

Fig. 5. Age range of chert samples, amout of magnetic microspherules collected, and computed mass influx rate during Permian and Triassic. Radiolarian assemblages are referred to YAO *et al. (1980) and* ISHIGA *(1986).*

mathematical formula:

$$M=1.96\times 10^{7} vV/m(\text{t/day}\cdot\text{earth}),$$

where v is the rate of chert sedimentation (mm/1000 yr), m, weight of the powdered chert (g), V, total volume of magnetic microspherules in a specimen (cm^3), and 1.96 as constant. This constant has been derived from the density of magnetic microspherule (taken as 5.2 g/cm^3), the density of chert (2.7 g/cm^3), the area of the earth ($5.101\times 10^{18} cm^2$), and the duration of an year (365 days).

The estimated rate of chert sedimentation is 2.8 mm/1000 yr for Late Triassic in Inuyama area (as computed from the data from YAO *et al.*, 1980) and 0.75 mm/1000 yr for Permian chert in Ryozen area (ISHIGA, 1986). The former is suited for the specimens of Inuyama and the latter for the specimens from Gujohachiman and Ryozen for calculations. The results are shown in Fig. 5. They indicate that the magnetic microspherules fell on the earth at the rate of about 1 t/day during Middle to Late Permian, and about 3 t/day during latest Permian to Middle Triassic.

However, this calculation may not be entirely accurate. The rate of sedimentation for Middle Triassic was extrapolated from that of the data for Late Triassic. Also, the estimates on the rate of chert sedimentation may not also be precise. If the same rate of the chert sedimentation applies to every specimen, then the results are almost similar values, except sample 21–29 and 18–3, which are near to zero, and the latest Triassic sample 21–1, which is three times more than the other values. Hence we treat the spherule sedimentation values obtained through the above procedure only as tentative. MURRELL *et al.* (1980) collected 22 mg of stony microspherules and 50 mg iron microspherules from deep sea clays in Pacific and calculated the mass influx rate of stony microspherules on the earth to be 90 t/yr. Judging from the relative amount of iron microspherules to stony microspherules collected by them, their result translates into 0.56 t/day fall of iron microsherules on the earth. It is interesting that there is an obvious approximation between the values obtained for the magnetic microspherules from Permian and Triassic with the above estimates.

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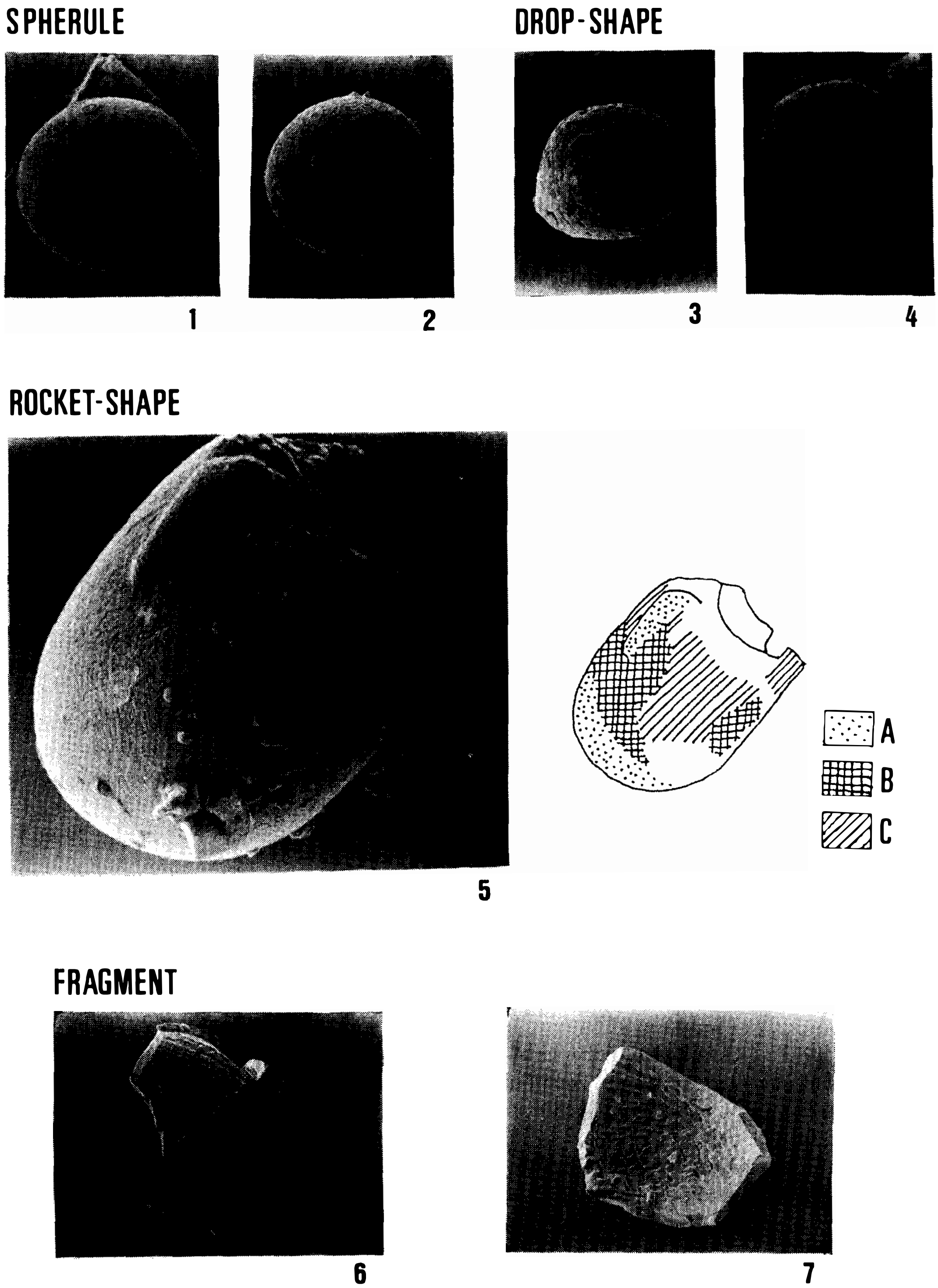

Plate 1. Spherule types.

		No.	Diameter (μm)			No.	Diameter (μm)
Spherical shape	1.	18-6-231	23.3	*Rocket-shape*	5.	20-0-1	84.0 × 66.0
	2.	18-6-105	21.3	*Fragment*	6.	20-0-52	44.9 × 29.5
Drop-shape	3.	18-6-99	21.0 × 18.3		7.	18-6-165	43.7 × 35.4
	4.	18-6-149	21.3 × 18.7				

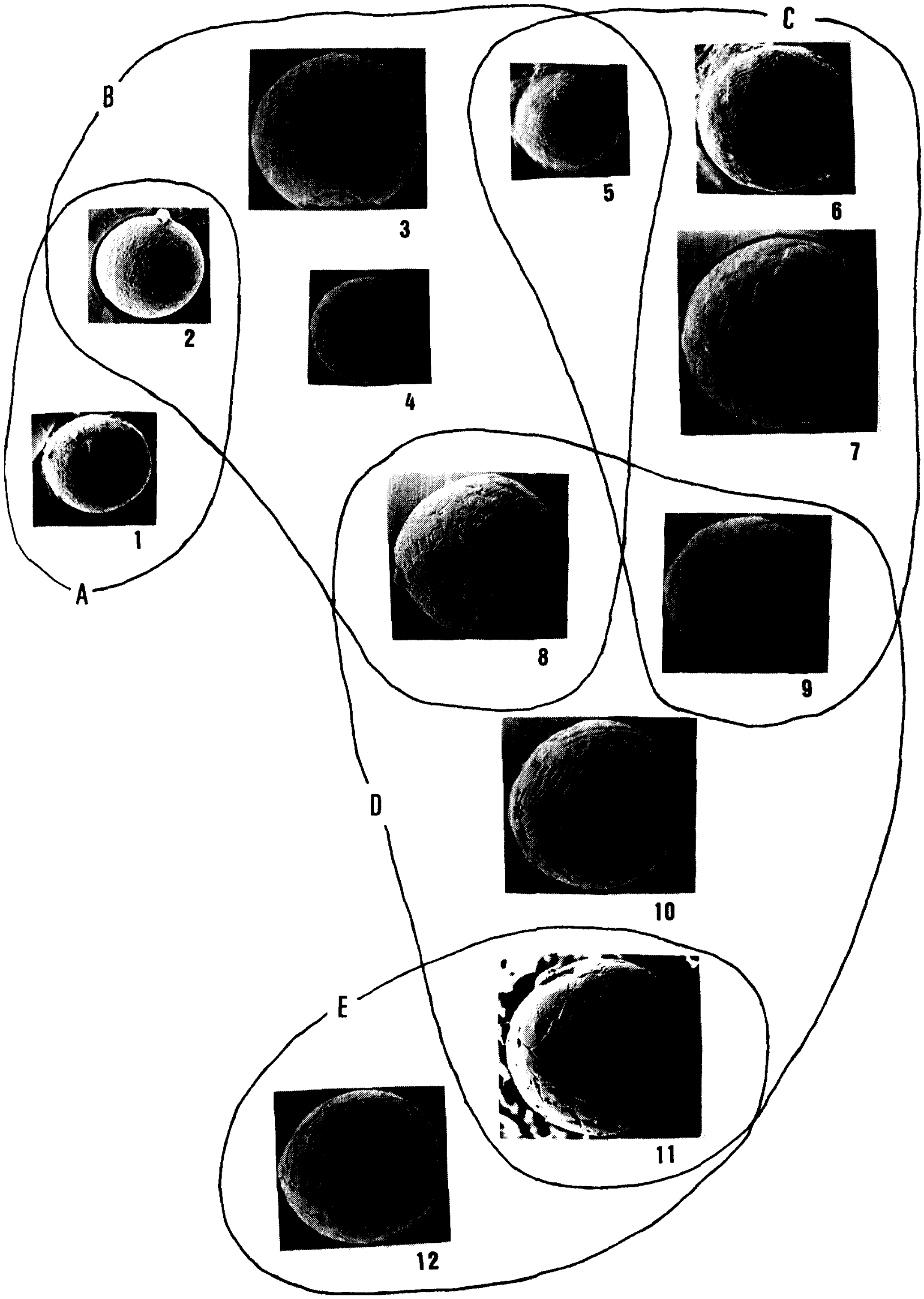

Plate 2. A to E types of microspherules.

	No.	Diameter (μm)		No.	Diameter (μm)		No.	Diameter (μm)
1.	3-8-14	16.0	5.	8-8-118	16.0	9.	18-6-260	24.0
2.	9-2-135	16.0	6.	3-8-131	34.0	10.	20-0-17	26.7
3.	18-6-244	25.3	7.	21-1-105	35.6	11.	9-2-1	84.0
4.	20-0-21	16.7	8.	21-1-6	50.7	12.	20-0-3	24.0