

## CHEMICAL COMPOSITION OF "COSMIC SPHERULES" IN MARINE SEDIMENTS

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### ABSTRACT

Metallic, silicate and glassy spherules were found in a beam-trawl net during the deep-sea expedition. Metallic and silicate spherules were analyzed chemically using EDTA titration, colorimetry and gravimetry. The chemical composition of metallic spherules agrees with that of the silicate phase in meteorites, while that of silicate spherules is similar to that of igneous rocks.

During the cruise of HMS Challenger John Murray discovered metallic spherules in deep-sea sediments and deduced in 1876 their extraterrestrial origin (1). After that discovery the "cosmic spherules" found in marine sediments has been studied by many workers (2)~(15). But the chemical composition of "cosmic spherules" has been determined individually using an x ray microanalysis (6)~(13) and a neutron activation analysis (14), (15).

The experimental data of individual spherules are, however, not so available to be applied as the chemical compositions of "cosmic dusts in space" in theoretical problems. Therefore the chemical determinations of average composition of "cosmic spherules" may be able to supply useful informations in space sciences.

In this work a number of spherules were analyzed chemically by means of EDTA titration, colorimetry and gravimetry method.

During the cruise\* of the Hakuho-Maru a beam-trawl net swept the pelagic deep sea bottom surface from the location (17° 16.9'N, 176° 18.2'W) to the location (17° 18.2'N, 176° 21.2'W) in the Mid-Pacific Mountain Region. In the beam-trawl net ferromanganese nodules and angular gravels or cobbles covered with red clay were obtained about 2 tons by weight.\*\* The red clay was washed out from them and the muddy water was filtered through sieves of 200 meshes. From the residual fine sand grains a number of spherules (metallic, silicate and glassy) with various diameter ranging 0.07~0.8 m/m were picked up with a fine needle under a view of microscope.

Metallic spherules possess ferromagnetic properties. They look like iron droplets, as which were boiled out from the heated surfaces of meteorites and then

\* This cruise was undertaken from December 1967 to February 1968 in the northern and western part of the North-Pacific Ocean with the "Hakuho-Maru", a research vessel of Ocean Research Institute, University of Tokyo.

\*\* Sample code; Station-40 (KH-67-5), Depth 4400 m.

cooled and solidified suddenly in the water. Some craters are found in the surface of several metallic spherules. (Fig. 1) A few "gourd-shaped" droplets are also found, which suggests that they have not been made up from larger bodies into such a shape and size on the deep sea bottoms. (Fig. 2).

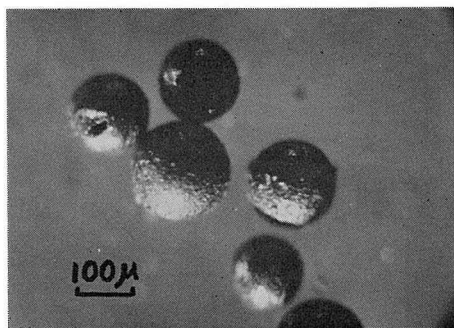


Fig. 1.

Fig. 1. Metallic spherules. In the upper left a crater can be seen.



Fig. 2.

Fig. 2. The "gourd-shaped" metallic spherules.

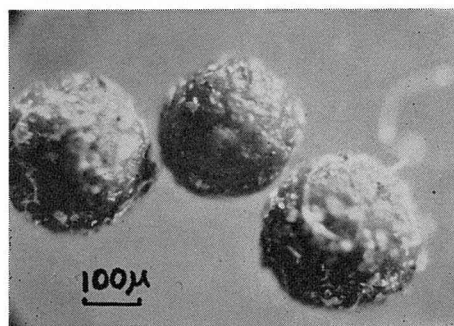


Fig. 3.

Fig. 3. Silicate spherules.

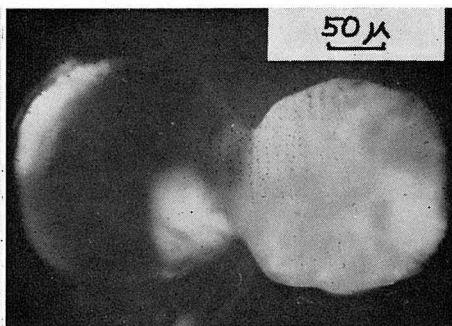


Fig. 4.

Fig. 4. Glassy spherules. The left; transparent, the right; semi-transparent.

Silicate spherules are of brown or yellow-brownish colour and they look just like "meat-balls". (Fig. 3) Several spherules are found wearing broken black-shinning crusts. Two glassy spherules are also found, the one is transparent and the another is semi-transparent. (Fig. 4)

Iron, nickel and manganese fractions of metallic spherules could be determined quantitatively using colorimetry respectively. The analyzed sample included 32 metallic spherules which were less than 100 μ in size. And it was 16.0 mg by weight. The results are shown in Table 1.\*

Table 1. The chemical composition of metallic spherules.  
(wt. %)

Fe	Ni	Mn
82.6	0.14	0.50

\* The chemical analysis of the spherules was carried out under our direction at Japan Analytical Chemistry Research Institute.

The chemical composition of silicate spherules were determined using gravimetry (Si, Al+Fe), EDTA titration (Fe, Mg+Ca, Ca) and colorimetry (Mn, Ni, Co, Cu). The results are shown in Table 2.\*

Table 2. The chemical composition of silicate spherules.

Si	Al	Mg	Ca	Mn	Fe	Ni	Co	Cu
24.03	9.53	0.45	0.58	0.04	0.96	0.014	0.020	0.015

The analyzed sample included more than 200 silicate spherules which were less than 100  $\mu$  in size. And it was 145 mg by weight.

In the Fig. 5 the chemical composition of metallic and silicate spherules are compared with the typical geo- and cosmochemical materials.

The manganese, iron and nickel fractions of the metallic spherules agree with

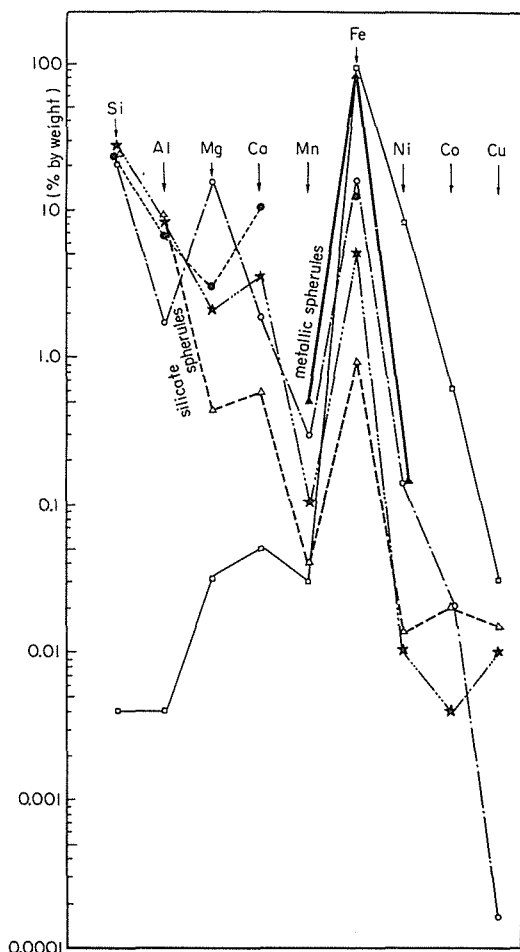


Fig. 5. The chemical composition of spherules compared with those of geo- and cosmochemical materials in grouping of "lithophile" elements (Si, Al, Mg, Ca, Mn), "siderophile" elements (Fe, Ni, Co) and "chalcophile" element (Cu).

- - - ○ : Silicate phase of meteorites (H. Brown) (16);
  - - - □ : Metal phase of meteorites (H. Brown) (16);
  - ★ - - ★ : Igneous rocks (V. M. Goldschmidt) (17);
  - - - ● : Lunar surface (Surveyor V~VII) (18)~(20);
  - ▲ - - ▲ : Metallic spherules ;
  - △ - - △ : Silicate spherules ;
- } present work KH-67-5, St-40.

\* The chemical analysis of the spherules was carried out partly under our direction at Japan Analytical Chemistry Research Institute.

those of the silicate phase in meteorites (16), while the chemical composition of the silicate spherules is similar to the average composition of igneous rocks (17).

In order to discuss the extraterrestrial origin of the "cosmic spherules" the detections of cosmic ray produced nuclides are necessary. In authors' laboratory the detections of radionuclides ( $Al^{26}$  etc.) are in preparation.

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