Nuclear Engineering and Technology 48 (2016) 293–298

Available online at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Invited Article

Origin of Spherule Samples Recovered from Antarctic Ice Sheet—Terrestrial or Extraterrestrial?

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ARTICLE INFO

Article history:
Received 7 February 2016
Accepted 12 February 2016
Available online 27 February 2016

Keywords:
Antarctic
Extraterrestrial
Iridium
Neutron Activation Analysis
Spherule

ABSTRACT

Thirty-eight spherules from the Antarctic ice sheet were analyzed using neutron activation analysis under two different conditions to investigate their origin. In almost all of these spherules, the contents of iron, cobalt, and manganese were determined to be 31% to 88%, 17 mg/kg to 810 mg/kg, and 0.017% to 7%, respectively. A detectable iridium content of 0.84 mg/kg was found in only one spherule, which was judged to be extraterrestrial in origin. A comparison of elemental compositions of the Antarctic spherules analyzed in this study with those of deep-sea sediment spherules and those of terrestrial materials revealed that most of the Antarctic spherules except for the sample in which iridium was detected could not be identified as extraterrestrial in origin.

1. Introduction

The *Challenger* expedition detected spherule samples for the first time from deep-sea sediments in the Pacific Ocean, some of which were reported to be extraterrestrial in origin [1]. Since then, the origin of those samples has been investigated in terms of their chemical compositions [2–4]. Spherule samples were found in the Antarctic and Greenland ice sheets as well as in deep-sea sediments [5]. Although many terrestrial and artificial contaminants were also detected in the Antarctic ice sheet, particles with matrix element compositions similar to those of the Allende and Murchison carbonaceous chondrites were separated from terrestrial contaminants by using a scanning electron microscope with an energy dispersive X-ray spectrometer [6]. Those particles, which included spherical and irregular shapes, are defined as Antarctic micrometeorites [6]. The measurement of noble gas compositions revealed that these Antarctic micrometeorites are extraterrestrial in origin and originated from carbonaceous chondrites [7,8]. We measured the chemical compositions of spherule samples recovered from the Pacific Ocean sea sediments through instrumental neutron activation analysis (INAA) by using the

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http://dx.doi.org/10.1016/j.net.2016.02.004

Kyoto University Reactor (KUR), and we designated spherules with relatively high iridium contents of 0.37–38 mg/kg as extraterrestrial [9,10]. In this study, the chemical compositions of 38 spherules from the Antarctic ice sheet were determined using INAA. In terms of both siderophile and lithophile element contents, the differences between the 38 Antarctic spherules and spherules from sea sediment measured in our previous study [9–11] were revealed. Additionally, whether the origin of the Antarctic spherules is terrestrial or extraterrestrial is discussed on the basis of the criteria used by Kobayashi and Ebihara [4] and Sekimoto et al [9] to judge the origin of spherules.

2. Materials and methods

The 38 spherule samples measured in this study were detected through microscopy from precipitated particles in a water tank at Dome Fuji station in Antarctica. The precipitated particles were F97B series supplied by the National Institute of Polar Research. By using a separation method based on grain size, magnetism, and other parameters [6], the F97B series were separated from the residue identified as 971127-2 by the National Institute of Polar Research after filtering the contents of the water tank. The diameters and weights of the 38 spherule samples ranged from several tens to hundreds of microns and a few to a few tens of micrograms, respectively. The elemental compositions of the spherules were analyzed with INAA under two different conditions using the KUR: (i) 50-minute irradiation using the pneumatic transport system (Pn-2) with a thermal neutron flux (n_{th}) of 2.75×10^{13} cm^2/s [12] and (ii) 50- to 70-hour irradiation at the inner reactor site with an n_{th} of 4.65×10^{13} cm^2/s. For the type ii irradiation, each spherule sample was washed with ethanol and then sealed in an aluminum foil bag. The spherule samples and a reference standard were packed in an irradiation capsule made of

Table 1 – Measured contents of the 38 Antarctic spherules evaluated in this study.

Sample	Weight (μg)	Fe (%)	Co (mg/kg)	Ni (%)	Ir (mg/kg)	Sc (mg/kg)	Mn (%)	Sb (mg/kg)	Co/Fe × 10^{-3}
1 spherule: Co/Fe ratio > 2.0 × 10^{-3}									
F97BN032	5.1	31.3 ± 1.5	675 ± 24	1.60 ± 0.15	0.84 ± 0.22	13.8 ± 1.4	0.237 ± 0.008	–	2.2
15 spherules: 0.085 × 10^{-3} < Co/Fe ratio < 2.0 × 10^{-3}									
F97BN005	6.0	62.7 ± 2.0	104 ± 13	<0.22	<0.03	4.5 ± 0.9	0.221 ± 0.003	<1.51	0.17
F97BN009	4.2	61.9 ± 4.4	807 ± 80	<0.70	<0.02	(0.13 ± 0.06)	0.594 ± 0.007	0.45 ± 0.09	1.3
F97BN010	4.5	65.5 ± 3.6	113 ± 33	<0.53	<0.92	<3.6	0.092 ± 0.002	–	0.17
F97BN011	2.3	80.8 ± 4.6	1,480 ± 86	<0.78	<0.04	0.69 ± 0.11	0.742 ± 0.010	1.5 ± 0.8	1.8
F97BN012	5.5	63.5 ± 2.8	229 ± 31	<0.34	<0.03	<0.19	0.419 ± 0.005	4.50 ± 0.95	0.36
F97BN013	10.0	65.5 ± 3.2	541 ± 41	<0.36	<0.04	<0.23	0.430 ± 0.005	1.7 ± 0.8	0.83
F97BN014	17.1	62.3 ± 1.4	634 ± 12	(0.08 ± 0.04)	<0.01	(0.11 ± 0.04)	0.429 ± 0.004	1.13 ± 0.27	1.0
F97BN017	3.2	76.6 ± 3.5	119 ± 34	<0.48	<0.06	<0.40	0.387 ± 0.006	4.88 ± 1.17	0.16
F97BN024	2.5	56.1 ± 4.2	568 ± 74	<0.74	<0.03	2.1 ± 0.2	0.516 ± 0.007	0.30 ± 0.08	1.0
F97BN027	10.4	41.3 ± 2.0	108 ± 17	<0.24	<0.02	(0.11 ± 0.06)	0.108 ± 0.004	<0.47	0.26
F97BN029	6.0	74.7 ± 2.4	1,530 ± 52	<0.28	<0.04	<0.29	0.348 ± 0.004	<1.83	2.0
F97BN030	5.3	39.4 ± 3.1	<45	<0.55	<0.03	25.2 ± 3.9	7.166 ± 0.163	<2.24	<0.11
F97BN036	5.0	67.3 ± 3.8	155 ± 32	<0.46	<0.05	<0.32	0.017 ± 0.001	<1.94	0.23
F97BN107	18.4	71.9 ± 5.5	63.4 ± 14.0	<0.10	<0.03	<0.19	0.326 ± 0.009	2.72 ± 0.19	0.088
F97BN116	3.0	69.0 ± 4.3	119 ± 34	<0.71	<1.25	<4.5	0.265 ± 0.009	–	0.17
22 spherules: Co/Fe ratio < 0.085 × 10^{-3}									
F97BN008	11.2	86.2 ± 3.5	37.3 ± 12.1	<0.27	<0.39	(1.8 ± 1.0)	–	–	0.043
F97BN015	8.0	64.9 ± 2.7	38.1 ± 10.6	(0.17 ± 0.11)	<0.02	<0.11	0.598 ± 0.022	1.63 ± 0.45	0.059
F97BN022	6.5	88.0 ± 4.1	(44 ± 19)	<0.40	<0.03	1.12 ± 0.12	0.324 ± 0.010	<0.86	0.050
F97BN026	12.8	77.5 ± 2.9	<17	<0.22	<0.02	<0.12	1.034 ± 0.026	1.42 ± 0.30	<0.022
F97BN028	18.6	83.0 ± 2.9	45.0 ± 8.5	0.17	<0.01	0.19 ± 0.06	0.534 ± 0.012	0.25 ± 0.12	0.054
F97BN035	10.1	81.9 ± 3.4	68.8 ± 16.0	<0.28	<0.41	<1.9	–	–	0.084
F97BN031	18.2	68.4 ± 2.6	50.1 ± 9.4	<0.18	<0.23	(1.8 ± 0.7)	0.382 ± 0.016	–	0.073
F97BN033	12.6	63.2 ± 2.8	(23 ± 11)	<0.23	<0.01	<0.07	0.323 ± 0.009	0.56 ± 0.16	0.036
F97BN101	24.0	69.3 ± 5.1	35.8 ± 8.0	<0.03	<0.02	(0.15 ± 0.07)	0.226 ± 0.006	0.25 ± 0.05	0.052
F97BN102	34.3	65.9 ± 1.6	40.6 ± 3.1	(0.08 ± 0.03)	<0.01	0.22 ± 0.09	0.342 ± 0.010	6.03 ± 0.32	0.062
F97BN103	21.7	78.0 ± 5.6	18.9 ± 4.0	<0.10	<0.02	1.5 ± 0.4	0.146 ± 0.004	0.88 ± 0.12	0.024
F97BN104	32.6	72.2 ± 1.8	15.7 ± 2.5	<0.07	<0.09	<0.50	0.073 ± 0.003	–	0.022
F97BN106	26.9	69.6 ± 1.8	50.6 ± 5.1	<0.09	<0.02	(0.12 ± 0.07)	0.111 ± 0.003	3.99 ± 0.23	0.073
F97BN110	16.1	67.9 ± 2.3	55.3 ± 9.3	<0.18	<0.02	0.36 ± 0.06	0.311 ± 0.009	3.87 ± 0.22	0.081
F97BN111	52.0	66.5 ± 4.8	42.1 ± 4.0	<0.02	<0.02	<0.14	0.262 ± 0.008	1.76 ± 0.14	0.063
F97BN112	29.1	71.3 ± 1.8	12.9 ± 3.5	<0.09	<0.12	<0.62	0.299 ± 0.009	–	0.018
F97BN113	14.6	71.0 ± 5.1	39.5 ± 10.0	<0.08	<0.03	<0.20	0.336 ± 0.010	1.28 ± 0.17	0.056
F97BN114	11.7	64.5 ± 2.1	(17 ± 7)	<0.18	<0.28	2.09 ± 0.67	1.274 ± 0.037	–	0.026
F97BN115	8.9	70.4 ± 5.6	53.4 ± 18.0	<0.25	<0.03	<0.26	0.365 ± 0.011	<0.35	0.076
F97BN117	5.7	68.1 ± 5.3	37.9 ± 17.0	<0.12	<0.13	<1.5	0.065 ± 0.003	–	0.056
F97BN119	3.0	67.9 ± 6.7	<45	<0.37	<0.24	<2.9	0.240 ± 0.009	–	<0.066
F97BN120	4.3	63.9 ± 2.4	(40 ± 16)	<0.31	<0.04	<0.28	0.231 ± 0.008	<0.40	0.063

–, Not measured; ( ), values are near the detection limit.