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Detail analysis of acid residues from various meteorites has been continued for study of the evolution of the solar system and information on processes of nucleosynthesis in the pre-solar stage. Last year we reported the results of chemical compositions of acid residues from three meteorites [Canyon Diablo (IA), Allende (CV3), Nuevo Mercurio (H5)] and the preliminary results of Ru isotopic compositions[1]. In these samples, Cr isotopic analyses were also performed to investigate 1) ^{54}Cr(neutron-rich e-process) anomaly and 2) the effect of extinct radioactive nuclide (^{53}Mn→^{53}Cr). The following progress report deals with the chemical and textural properties of acid residues from Murchison(CM2), La Criolla(L6), Qingzhen(E3) and Cr isotopic compositions measured so far.

Major element compositions in acid residues of Murchison, La Criolla, Qingzhen are shown in Table1 along with the fraction of these acid residues. Representative SEM photographs with EDS spectra of the aliquants of acid residues of them are shown in Fig. 1. It is known that spinel, hibonite, chromite etc. as well as carbonaceous matter are present in Murchison residues [e.g. 2], and in our sample, these minerals may be included. But high S content indicates that it contains much S-bearing particles and/or organic matter or something. On the other hand, acid residues of La Criolla consist of chromite for the most part judging from abundant Cr and Fe contents by INAA and Cr:Fe ratios of about 2:1 in the EDS spectra. Ti and V are also much enriched (Ti: 1.51%; 35 times/CI; V: 0.445%; 79 times/CI) in La Criolla residues. In acid residues of Qingzhen, Cr content is relatively low than that of other meteoritic residues. It is consistent with the fact that enstatite chondrite contain no chromite[3]; and Cr:Fe ratios of about 2:1 in the EDS spectra and fairly abundant S content show that daubreelite may be present in Qingzhen residues. In addition, enrichment of refractory siderophiles is characteristics of acid residues for any kind of meteorites. We are now analyzing TEM bright field images and SAD(selected area diffraction) patterns of these acid residues to idetify minerals therein definitely.

The samples (HCl/HF residues) were decomposed in sealed teflon vessels by a microwave dissolution method with several mixed acids and Cr was separated by precipitation and anion exchange method[4]. In mass spectrometric techniques of Cr, the zone-refined (99.995%) outgassed V-shaped Re single filament with silica gel and boric acid was employed. Isotopic analyses were performed by a VG 354 thermal ionization mass spectrometer.

Preliminary Cr isotopic analyses were carried out for acid residues of Allende, Nuevo Mercurio and La Criolla so far. Remarkable isotopic anomalies far beyond experimental errors could not be detected for both ^{53}Cr/^{52}Cr and ^{54}Cr/^{52}Cr, but in acid residues of Allende, ^{54}Cr/^{52}Cr are slightly enriched relative to normal values though errors are rather large. In recent paper, Rotaru et al. [5] suggested that Cr isotopic composition of the Solar System results from the mixing of several major components with distinct isotopic compositions, and that particularly C1 and C2 chondrites keep, at least partially, the memory of their initial isotopic diversity. We expect such heterogenities could be detected in our various kind of samples. Isotope abundance studies in acid residues, concentrating on more specific mineral phases by means of stepwise dissolution method, are currently underway.

Aknowledgements -- We thank Y. Suzuki of Yamagata University for analyses of samples by TEM/SEM-EDS.

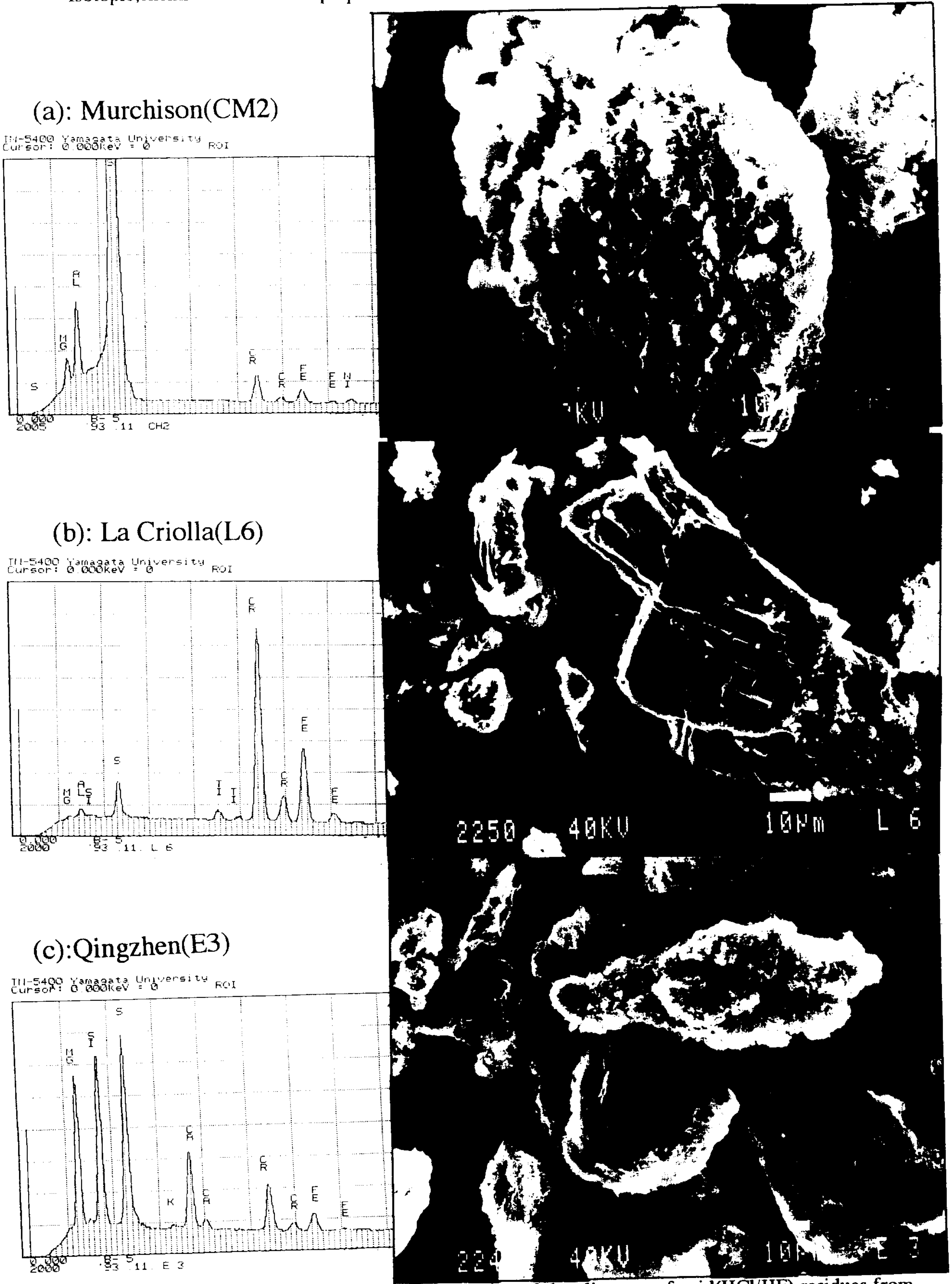
References : [1] Kano, N. et al. (1993) LPS(Abstracts), **24**, 749 [2] Alaerts, D. N. et al. (1980): GCA, **44**,189 [3] Srinivasan, K. et al. (1977): JGR, **48**, 2567. [4] Lee, T. and Tera, F. (1986): GCA, **50**, 199. [5] Rotaru et al. (1992): Nature, **26**, 29-36.

Table 1 Major element contents in acid residues of three meteorites

Sample		Acid residues	combustion method(%)				INAA$^{\#}$ (%)				
		fraction (wt.%)	C*	H*	N*	S*	Cr	Fe	Ni	Al	Mg
Murchison(CM2)	HCl/HF residue	3.12	34.6	2.10	1.48	46.5	1.14	0.694	0.240	0.958	0.943
	HCl/HF-HNO$_3$ residue	1.30	29.5	1.71	2.56	27.5	1.75	3.50	0.191	1.13	2.15
La Criolla(L6)	HCl/HF residue	0.83	1.20	0.16	---	12.8	35.6	18.8	0.424	2.76	2.75
Qingzhen (E3)	HCl/HF residue	0.65	6.26	2.05	0.71	7.27	0.409	0.800	0.0216	0.673	7.92

*) We are indebted to T. Seki of Tokyo University for determing C,H,N,S.contents of samples.

#) Errors of INAA data are below 5% except Ni in Qingzhen(~10%). Neutron irradiations and use of the counter facilities were carried out at the Inst. for Atom. Energy Res. in Rikkyo University.



(a): Murchison(CM2)

(b): La Criolla(L6)

(c):Qingzhen(E3)

Fig. 1 Reprentative SEM pictures with EDS spectra of the aliquants of acid(HCl/HF) residues from three meteorites((a): Murchison(CM2), (b): La Criolla(L6), (c):Qingzhen(E3)).

COSMOGENIC ^{26}Al IN DEEP-SEA STONY SPHERULES

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Size dependence of averaged cosmogenic ^{26}Al activity in deep-sea stony spherules were estimated considering the effects of orbital evolutions by the Poynting-Robertson effect in the interplanetary space and the atmospheric entry. It is found that the contribution of the dust particles with high eccentricity in interplanetary motion can be ignored practically because such particles would have so high atmospheric entry velocities that they would disappear in the upper atmosphere besides the probabilities of capturing by the earth are low. To examine the results by measurering ^{26}Al activities in the spherules, we expect, the Poyinting-Robertson effect can be verified.

Radionuclides ^{26}Al is produced in the interplanetary dust particles through irradiation of the solar cosmic rays. The amounts of such nuclide production would have been determined by the motion of the dust particles and the spatial intensity of the solar cosmic rays. The Poynting-Robertson effect which has been thought to be decisive for the motion of the interplanetary dust particles has been not yet proved observationally and also experimentally. In reality, the interplanetary dust particles have various origins and are experienced complex processes of evolution. For example, comets scatter the dust near the sun which are thought to be main source of the dust around the earth's orbit. And the collisional process of the dust particles is also important. Then, it is not so clear whether the dust particles which have been affected mainly the Poynting-Robertson effect only are present or not. We think that the deep-sea spherules are the candidates of that,which would be proved by measuring the long-lived cosmogenic radionuclides in them.

In the following calculations, changes of the orbital elements a (semimajor axis) and e (eccentricity) of the paticle on an elliptical orbit according to the Poynting-Robertson effect[1] are taken into account. The energy spectrum of the solar cosmic rays, cross sections of ^{26}Al producing reactions, and p/α ratio were to be followed Lal and Venkatavaradan[2]. And the spatial distribution of the solar cosmic ray intensity is to be inversely proportional to r (heliocentric distance). The starting point was set to $a = 2.8$ *AU* which is the average heliocentric distance of asteroids. The results would be not so differed in the case of the more distant starting points due to the character of the spatial distribution of the solar cosmic ray intensity.

Fig. 1 shows ^{26}Al cumulation with time in the particle of 100μm in diameter moving from $a = 2.8$ *AU* to $a = 1.0$ *AU* according to the Poynting-Robertson effect with each initial eccentricity e_0. In the case of $e_0 < 0.3$, ^{26}Al production is almost equal to that in the case of circular orbit. In the case of higher eccentricity, however, ^{26}Al production decreases.

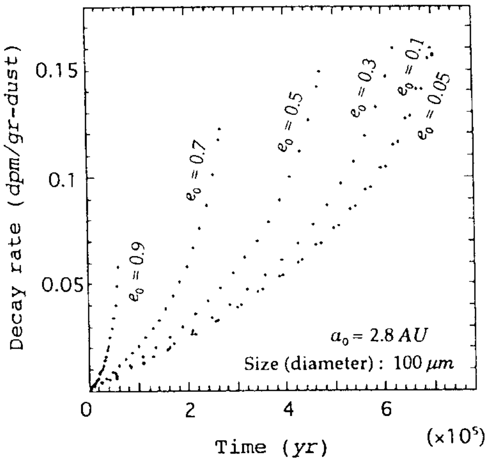

Fig. 1. ^{26}Al cumulation with time in the particle moved from $a = 2.8$ *AU* to $a = 1.0$ *AU* according to the Poynting-Robertson effect with each initial eccentricity.

A particle having low eccentricity will be trapped by the earth when the semimajor axis a is nearly equal to 1 *AU* Hence, we expect for ^{26}Al production in such particle the value of the top point of the curve drawn in Fig. 1. But in the case of high eccentricity, a particle would cross the earth's orbit even when it's semimajor axis had not yet approached 1 *AU*. So we must calculate the probability for the particle to be captured by the earth in each stage of orbital evolution to estimate the average nuclide production, and the atmospheric entry velocity in each case to take the effect of the atmospheric entry into account.

Fig. 2 shows the expected atmospheric entry velocity of particles in the case of the initial eccentricity $e_0 = 0.1$, 0.3, 0.5, 0.7 and 0.9 with 0.1 *rad.* of the inclination of the orbial plane. In the low eccentricity case, the particle captrue probability is high and the entry velocity is low. In the high eccentricity case, particularly $e_0 > 0.7$, the particle would rarely be captured by the earth and if captured, the entry velocity would be very high. By performing the atmospheric entry calculations[3][4], we can see that the process make the final sizes of particles be very small regardless of the initial sizes when the entry velocity is high. Fig. 3 shows the average ^{26}Al production in each entry velocity. In the high eccentricity cases, ^{26}Al productions are fairly low as compared with the low eccentricity cases, however, which would not make serious influences on the whole average of ^{26}Al. Moreover, the atmospheric entry process would further weaken the influences because of their high entry velocities.

Similar calculations were performed in various sizes, then atmospheric entry processes were calculated in each, and finally summed up the results to get the final size dependences of ^{26}Al productions. In this calculation, size distribution of the particles was to be of $dn \propto s^{-4}ds$ at the starting point in the interplanetary motion which agrees with the terrestrial observation and the experiments carried out on space crafts[5]. Actually, interplanetary dust particles would be the mixed ones of various eccentricities, so we must average various cases of the initial eccentricity e_0. In Fig. 4, the circles represent the average ^{26}Al productions of after-atmospheric entry of the cases of every 0.05 steps of e_0 below 0.5, and the crosses represent that of below 0.75. Fine dotted line represents the case of the circular orbit ($e_0 = 0.0$) of pre-atmospheric entry. That these two results of after-atmospheric entry are almost the same though there is some difference in pre-atmospheric ones, means that the cases of e_0 above 0.5 merely do contribute to the final results.

The influence of variation of the inclination of the orbital plane is not considered here, however high inclination cases do not contribute to the after-atmospheric entry results by the atmospheric filteration. It can be said that if the cases of various orbital elements are taken into account, the final size dependence of ^{26}Al productions does not differ seriously from the case of the circular orbit ($e_0 = 0.0$) of pre-atmospheric entry, which is easier to calculate.

References

[1] Wyatt, S. P. and F. L. Whipple (1950) Astrophys. J., 111, 134–141

[2] Lal, D. and V. S. Venkatavaradan (1967) Earth. Plan. Sci. Lett., 3, 299–310

[3] Matsuzaki H. and K. Yamakoshi (1993) Lun. Plan. Sci. (Abstracts), 24, 943-944

[4] Love, J. J. and D. E. Brownlee (1991) Icarus, 89, 26–43

[5] Grün, E. et al. (1985) Icarus, 62, 244–272

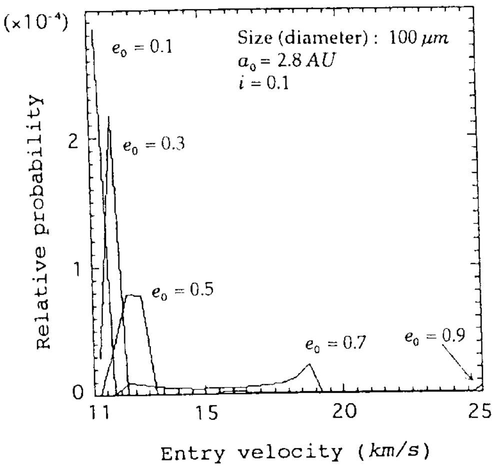

Fig. 2. Expected atmospheric entry velocity of particles in the case of the initial eccentricity e_0 = 0.1, 0.3, 0.5, 0.7 and 0.9.

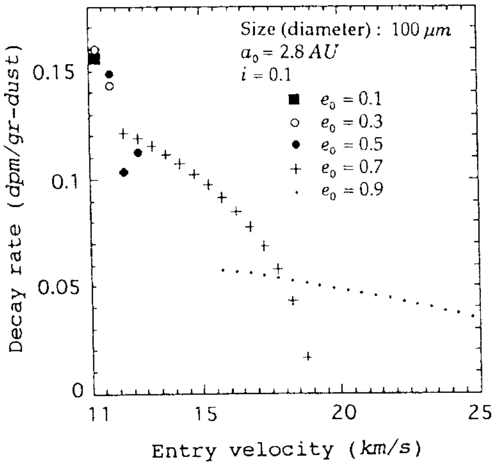

Fig. 3. Average ^{26}Al production in each entry velocity corresponding to Fig. 2.

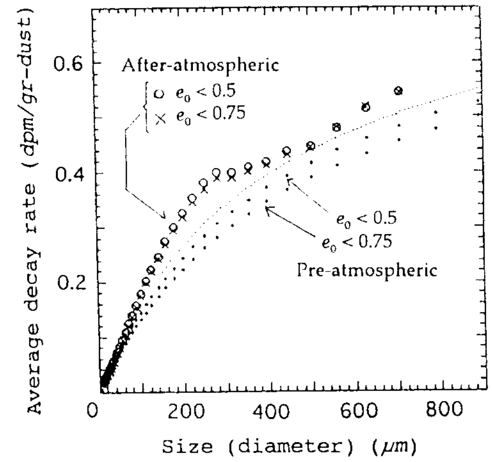

Fig. 4. ^{26}Al production of the average of each case of every 0.05 steps of e_0 below 0.5 and below 0.75. Fine dotted line represents the case of the circular orbit (e_0 = 0.0) of pre-atmospheric entry.

BROWNLEE'S PARTILES OF COSMIC ORIGINS IN DEEP SEA SEDIMENTS

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Recently Several particles have been discovered among strongly magnetized grains gathered from deep sea sediments, whose compositions are muchsimilar to those of Brownlee's particles (stratospheric) of the category "C" in the Cosmic Dust Catalog compiled by CDPET of NASA. The used deep sea sediments were dredged by R/V Hakurei-Maru at off Hawaiian Isl-ands at a depth of 4500 m.

Suzuki et al.(1993) [1] have been searching extraterrestrial grains witha high sensitive x-ray microanalyzer among several hundred magnetc grains, whose sizes exceed 74 μm in the magnetic fraction. A picture ofthe samples is shown below;

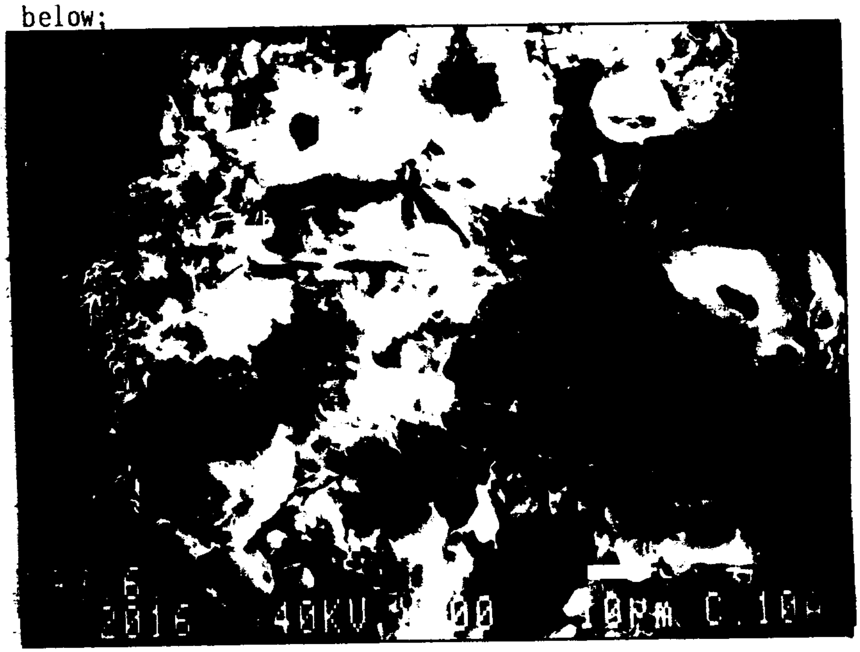

Fig.1 SEM(Scanning Electron Microscope) picture of a typical grain. The size is 100x90μm.

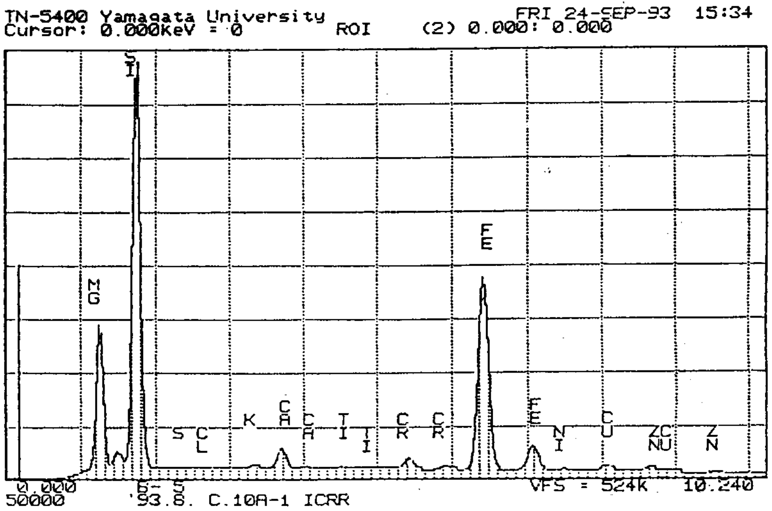

Fig.2 The diagram of the chemical composition of the grain shown in Fig.1. In the diagram Cu and Zn peaks are originated from base materials (brass) of the devices.

The samples were weighed with an electrobalance(Cahn #29, accuracy; 0.1μm). Preliminary results of major chemical compositions of a few grains, whose data were obtained using a TEM device.
The results were averaged with the data of 10 point measurements overallthe sample. (% by weight)

Sample Code	Weight (μg)	(Si)	(Fe)	(Mg)	(Ca)	(Cr)	(Ti)	(Ni)	(Al)	(S)
#A	3.8	38.7	32.2	20.9	2.2	1.8	--	--	--	--
#C	6.7	4.0	75.1	0.7	0.7	--	0.9	5.7	1.4	11
#D	1.8	1.8	81.5	--	0.7	--	4.0	5.3	3.5	--

The rare and noble metals are much enriched and the measurements were performed by INAA using Allende powder as a reference.
The samples are so small that the statiatical errors(± σ) were not better than 50～60% .

Sample Code	(Ir)	(Pt)	(W)	(ppm by weight)
#A	--	103	45	
#C	2.4	--	--	
#D	43	72	--	

(Significant values of Au contents were obtained, however,it is possible, Au-pollution could happen in vacuum devices of SEM and XMA.)

Up to now, fragile, unmelted and thermally-ungenerated grains of comic origins have been discoverd in the stratosphere [2] and arctic ice layers [3,4]. In this work large-sized, unmelted grains could be discovered from deep sea sediments, so that it is expected, some fruitful comparisons between deep sea spherules and deep sea Brownlee's particles [5].

REFERENCES

1 Suzuki Y.et al.(1993) presented at the Fall Meeting of the Jap. Soc. Planet.Sci.
2 Compiled by CDPET of NASA; Cosmic Dust Catalog.
3 Maurette M.et al.(1991) Nature 351, 44.
4 Koeberl C. et al.(1992) LPSC XXIII 709.
5 Yamakoshi K.(1993) EXTRATERRESTRIAL DUST,Kluwer Acad. Publ.