

The Chemical Composition, Petrography and Mineralogy of the Japanese Chondrite Nagai

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I. Introduction

On May 30, 1922, at about 10 a.m. local time, when three farmers were taking a rest together on a footpath between rice fields, they heard a detonation in the southwest. Soon after that, a stone fell into a water-covered rice field in front of them and another three stones flew over them rapidly. Immediately, one of them, the late T. Inoue picked up the stone, when it was still warm.

Fig. 1 shows the rice field into which the meteorite fell. The site is now in the city of Nagai, Yamagata-ken, $38^{\circ}07.3' N.$, $140^{\circ}03.7' E.$ The other three stones were unfortunately never recovered. The stone was named "Nagai" taking the present name of the place. The finder kept the stone very carefully and never showed it to the public. One of the authors, S. M., accidentally heard about it and recognized it to be a chondrite in May 1977. Fig. 2 shows one of the witnesses, H. Takaya holding the stone in front of the Inoue's house.

The complete mass of Nagai is shown in Fig. 3. The stone weighs 1.81 kg, and

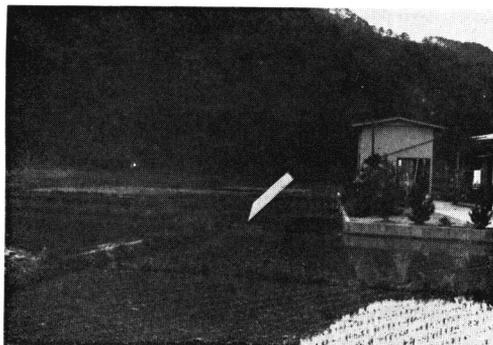


Fig. 1. The rice field in Nagai, Yamagata-ken, where Nagai chondrite fell



Fig. 2. One of the witnesses, H. Takaya holding the Nagai chondrite in front of Inoue's house.

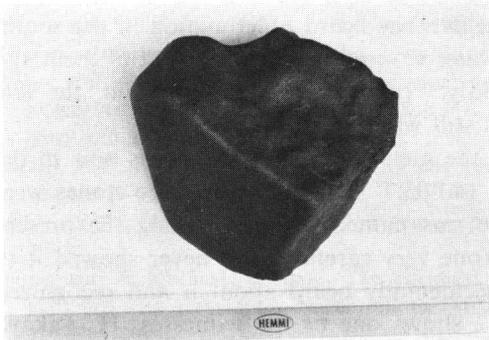


Fig. 3. The complete mass of Nagai chondrite

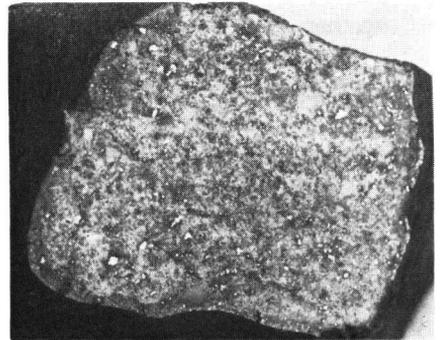


Fig. 4. Nagai chondrite. Freshly cut surface

is completely covered by black fusion crust. The main mass of Nagai is still in the possession of the Inoues, and they kindly provided us with a small end piece. The freshly cut surface is shown in Fig. 4. It is already somewhat oxidized, but showed typical chondritic structure. The black fusion crust is remarkably thick.

In this paper, the chemical composition as well as detailed petrology and mineralogy on Nagai are presented.

II. Chemical Composition of Nagai

A. *Experimental*

The chemical composition of Nagai was determined by a similar method to that employed in the determination of Yamato (a), (b), (c), (d), Numakai (Shima, 1974), and Brazilian meteorites (Shima *et al.*, 1974, Levi-Donati *et al.*, 1976). The method

comprises three kinds of sample decomposition procedures, that is, 1. HF-HClO₄ dissolution; 2. Na₂CO₃ fusion; and 3. stepwise chemical fractional dissolution. These are described briefly as follows:

1. *HF-HClO₄ dissolution*: This begins with a dissolution using the mixture HF+HClO₄, and is employed especially when analyzing for alkaline elements, as well as for most of the other major elements. About a 0.5 g chunk of the specimen was weighed and dissolved in the mixture of HF and HClO₄ after removing the outer crust. Both acids were used a little in excess over the equimolar quantities of major elements in the sample. After all excess HF was removed, the solution was made up to about 100 g with diluted HCl and weighed. An aliquot of the solution was taken by weight and diluted to an optimum concentration of each elements for determining the concentration of Mg, Al, Ca, Na, K, Cr, Mn, Fe, Ni, Co, Ti and Cu by atomic absorption spectrometry. Another aliquot was used for the main component and some of the trace element analyses by emission spectrometry after diluting the solution with water to 10 times. The emission spectrometry of the main component elements of Nagai, Mg, Ca, Al, Ti, Fe, Ni, Co, Mn, Cr and P and of the trace elements V, Cu, Zn, Sr and Zr, was performed by an emission spectrograph with an inductively coupled radio frequency plasma, model ICPQ-100, manufactured by Shimadzu Seisakusho Ltd. The detail function of the instrument and the procedure were described elsewhere (Imai *et al.*, 1978, Shima *et al.*, 1978).

Phosphorus was analysed by colorimetry (Ikeda 1951) after separating Fe and Co using an anion exchange procedure (Shima 1974).

2. *Na₂CO₃-KNO₃ fusion*: The main purpose of this method was to analyze for Cr, SiO₂ and S. About a 0.5 g chip of the Nagai specimen was fused with 4 g of Na₂CO₃ and 0.5 g of KNO₃. The fused melt was extracted with H₂O and the residue was

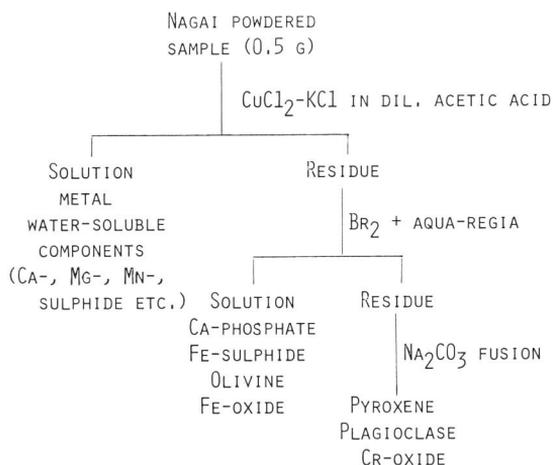


Fig. 5. Schematic diagram of the stepwise chemical fractional dissolution of Nagai chondrite.

dissolved in HCl. Silica was precipitated from both solutions by repeated evaporation of the solution to dryness and dissolution of the residue with diluted HCl. Silica was then determined gravimetrically. From the H₂O extract of the fused melt, S was determined gravimetrically in the form of BaSO₄. After separating the BaSO₄ precipitate, the solution was used for colorimetry of P and emission spectrometry of Cr. The HCl extract of the residue of fused melt was used for analyses of Mg, Ca, Al, Ti, Fe, Ni, Co, Mn and Cu by atomic absorption spectrometry and emission spectrometry.

3. *Stepwise chemical fractional dissolution*: This method was employed for the accurate measurement of S and the determination of the chemical state of Fe. The brief scheme used in this experiment is given in Fig. 5 and the method is summarized in Table 1. Detailed description and application of the method have appeared elsewhere (Shima, 1974, 1976). The main component elements such as Fe, Ni, Co, Mg, Al, Ca, Cr, Mn and Ti in each fraction were analyzed by atomic absorption and P was again determined colorimetrically.

Table 1. Details of the stepwise fractional dissolution method

Name of Fraction	Minerals mainly dissolved in the fraction	Composition of reagent	Treatment
CuCl ₂ -KCl	Metal Calcium sulphide & H ₂ O soluble Min.	30 ml 10%-2KCl- CuCl ₂ -2H ₂ O soln. +0.2 ml acetic acid	room temp. 30 min, in a continuous stream of CO ₂
Br ₂ + aqua-regia	Calcium Phosphate Iron sulphide Olivine Magnetite	3 ml Br ₂ in 25 ml aqua-regia	Agitate till vigorous reaction ceases, then warm up gradually until excess Br ₂ is evaporated
Residue	Pyroxene Plagioclase Chromite	Na ₂ CO ₃	fusion

B. Results

Analytical data obtained by the various methods are tabulated in Table 2. In Table 3, the results of the fractional dissolution method are shown. The data for Ni and Co are not 100% and show the same values of both elements in CuCl₂-KCl fraction. This could result from the incomplete dissolution of metal in Nagai into CuCl₂-KCl solution, or from the oxidation of metal which occurred during the storage of the chondrite in a private house. From this Ni and Co data, it is assumed that ~18% of metallic Fe is also not dissolved in CuCl₂-KCl solution. Iron in the Br₂ fraction should consist of part of the metallic, and all of the sulphide iron and the silicate iron in olivine. Sulphide iron, i.e. troilite iron, is calculated as equimolar amount of S.

CuCl₂-KCl solution dissolves 0.35% of Mg and 2.2% of Ca, other than Fe, Ni and Co. These elements seem not to be in metallic phase, and should be composed

Table 2. Examples of analytical results obtained by the various methods

Element (%)	Acid decomp.		Alkaline decomp.		others
	ICPQ-100*	A.A.**	ICPQ-100*	A.A.**	A.A.**
MgO	21.69	24.24	19.95	24.44	21.57
Al ₂ O ₃	2.29	1.96	1.83	1.96	1.99
Cr ₂ O ₃	0.10	0.12	0.38	0.66	0.68
MnO	0.36	0.34	0.28	0.31	0.37
P ₂ O ₅	0.37	0.308***	—	0.352***	0.345***
S	—	—	—	2.25****	2.55****
Fe (total)	21.82	21.36	20.37	20.44	21.05
Ni	1.27	1.13	1.19	1.22	1.10
Co	0.063	0.058	0.062	0.063	0.056
Cu (ppm)	91.4	96	82.5	123	—

* Emission Spectrometry

** Atomic absorption spectrometry

*** Colorimetry

**** Gravimetry

Table 3. Relative abundances from stepwise dissolution

Element	Fraction (%)		
	CuCl ₂ -KCl	Br ₂ +Aqua-regia	Residue
Mg	0.35	63.40	36.25
Al	~0	~0	100
Ca	2.2	21.0	76.8
Cr	~0	0.8	99.2
Mn	~0	55.7	44.3
Ti	~0	10.5	89.5
Fe	28.0	56.5	15.5
Ni	81.6	18.4	~0
Co	81.7	18.3	~0
P	~0	100	~0

of somewhat water-soluble minerals.

Al, Cr and Mn were not dissolved in CuCl₂-KCl solution. This indicates that the weathering of Nagai chondrite is not so serious as expected.

All phosphorus was dissolved in acidic Br₂ water and 99% of Cr remains in the residue. This may indicate that this chondrite was highly metamorphosed and recrystallized (Shima, 1974, Levi-Donati *et al.*, 1976).

From Mg and Fe in the Br₂ fraction and in the residue, one can calculate fayalite and ferrosilite in olivine and pyroxene respectively.

In Table 4, the chemical composition of Nagai is presented in conventional form.

For the comparison, the literature values of each element in various ordinary chondrite (Mason, 1971, Shima, 1979) are also tabulated. The sum of the abundance of the main component elements is less than 100%. This may have been caused by

Table 4. Chemical composition of the Nagai Chondrite and the literature values of ordinary chondrites*

(%)	Nagai	H	L	LL
SiO ₂	38.73	34.2 — 37.9	38.1 — 41.7	38.7 — 41.3
MgO	24.34	21.97—24.82	23.30—26.52	24.01—26.12
FeO	12.56			
Al ₂ O ₃	2.08	1.68— 2.12	1.89— 2.48	2.02— 2.27
CaO	1.83	1.61— 1.71	1.71— 1.89	1.50— 1.68
Na ₂ O	1.39	0.77± 0.09	0.88± 0.11	0.89± 0.11
K ₂ O	0.13	0.06— 0.13	0.07— 0.12	0.02— 0.18
Cr ₂ O ₃	0.57	0.50± 0.06	0.56± 0.06	0.54± 0.06
MnO	0.35	0.29± 0.22	0.32± 0.19	0.33± 0.13
TiO ₂	0.10 ₂	0.09— 0.13	0.08— 0.14	0.09— 0.22
P ₂ O ₅	0.32	0.23— 0.25	0.18— 0.25	0.18— 0.41
Fe	7.08			
Ni	1.23	1.21— 2.06	0.67— 1.57	0.39— 1.57
Co	0.060	0.04— 0.13	0.03— 0.09	0.02— 0.11
FeS	6.58	2.52— 7.21	2.14— 6.58	4.99— 7.10
Sum	97.35			
Total Fe	21.01	24.57—30.88	20.15—23.61	18.56—21.30
(ppm)	Nagai	Ordinary chondrites		
V	39.8	44 — 94		
Cu	91.4	80 ± 40		
Zn	85.7	8 — 102		
Sr	12.4	8 — 12		
Zr	10.2	4.9— 8.7**		

* These values are taken from the "Handbook of Elemental abundances in Meteorites" B. Mason Ed. 1971.

** Shima, 1979.

Table 5. Identification of Chemical Group

	Nagai	H	L	LL
Fe/SiO ₂	0.54	0.77±0.07	0.55±0.05	0.49±0.03
Fe (metal)/Fe	0.337	0.63±0.07	0.33±0.07	0.08±0.07
Fayalite ratio	24.8 25*	18±2	24±2	29±2
SiO ₂ /MgO	1.59	1.55±0.05	1.59±0.05	1.58±0.05

a little oxidation by the weathering. From Table 4, it is apparent that Nagai belongs to L group ordinary chondrite.

From Table 3 and Table 4, one can easily calculate the weight ratios of SiO₂/MgO, Fe(total)/SiO₂, Fe(metal)/Fe(total) etc. which have been suggested as indicator values of the chemical classification of chondrites by Van Schmus and Wood (1967). The results are shown in Table 5, together with the values presented by Van Schmus and Wood for each ordinary chondrite. The fayalite ratio calculated from the

chemical data in Table 3 agrees well with that of the estimated one by optics. This together with the results in Table 3 indicates that Nagai olivine is quite homogeneous, and this chondrite may lie in 5 or 6 of the petrologic class.

III. Petrography and Mineralogy of Nagai

A. Experimental Method.

Polished section and polished thin section of chondrite sample were prepared for microscopic investigation, and the mode of occurrence of minerals and the texture were investigated by the polarization microscope. The optic axial angle and optical orientation of non-opaque minerals were measured with the aid of a universal stage which was set on the microscope stage. The refractive indices of main silicate minerals, olivine, orthopyroxene and plagioclase, were determined by the oil immersion method using finely pulverized samples. The error is estimated to be ± 0.003 .

Cell dimensions of olivine and orthopyroxene were calculated from X-ray powder patterns taken by the D-9C X-ray diffractometer made by Rigaku Electric Co. Ltd., using graphite-monochromatized Cu $K\alpha$ radiation. The diffraction angles of peaks were corrected in comparison with those of pure silicon powder. The Debye method was also applied for the identification of mineral species. Crushed mineral grains mixed with standard silicon powder were mounted on the thin glass rod with an adhesive resin, and photographs were taken with a camera, 114.6 mm in diameter, under Fe $K\alpha$ radiation.

B. Results.

The main mineral species of Nagai are olivine, pyroxene, plagioclase, nickel-iron and troilite, and apatite and chromite are present in accessory amounts. The mineral composition obtained by point counting is shown in Table 6. Nagai exhibits a structure characteristic of a chondritic meteorite having suffered advanced recrystallization as seen in Fig. 6 (Dodd *et al.*, 1967), and is ascribed to an olivine-hypersthene chondrite of Prior's (1920) classification according to the average molar composition of olivine and orthopyroxene (Keil and Fredriksson, 1964; Mason, 1963).

Chondrules, deformed generally in shape, survive as relict, and their outline is usually indistinguishable in transmitted light. The relict of chondrule structure is

Table 6. Modal composition of Nagai chondrite.

volume %	Matrix	Chondrule
Total 100	84	16
Olivine	57	57
Orthopyroxene	25	30
Clinopyroxene	1	3
Plagioclase	11	5
Apatite	0.5>	—
Opaque minerals	7	4

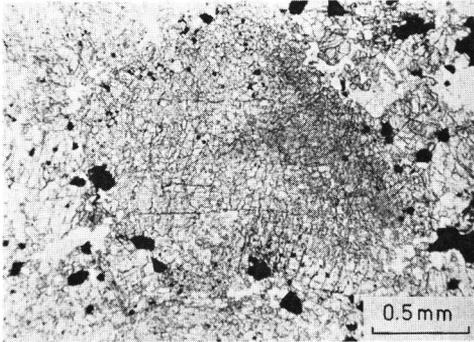


Fig. 6. An olivine chondrule in the matrix of Nagai chondrite. The shape is deformed, and the outline is poorly definable in the matrix.

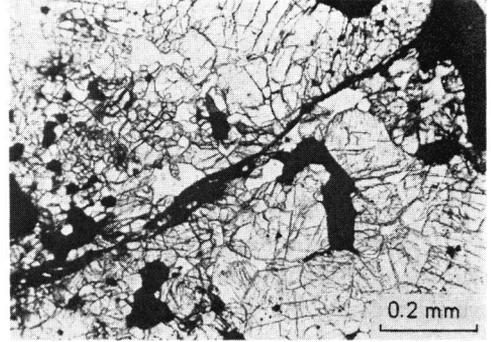


Fig. 7. A black vein penetrating the matrix of Nagai chondrite.

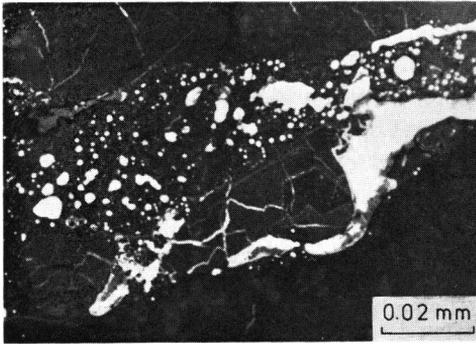


Fig. 8. A magnified view of the black vein under the reflected light. Fused globules of nickel-iron and troilite are observed in the vein.

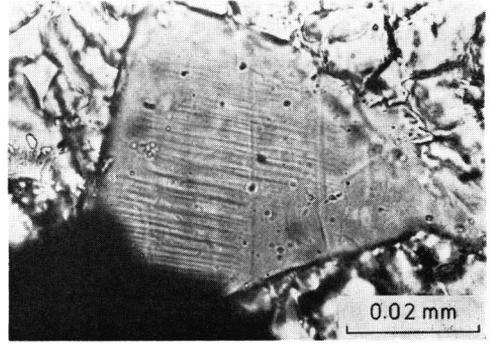


Fig. 9. Deformation lamellae observed in plagioclase near the black vein.

poorly discernible by the presence of parallel-layered or radial arrangement of elongated olivine and pyroxene crystals and by the rounded shape of mineral aggregation.

The matrix of this meteorite is granulated, and its silicate phase consists of a transparent and crystalline mass composed of olivine, pyroxene and plagioclase grains. Wavy extinction is prevalently observed in every silicate grain under cross polarized light. In the local part of thin section, thin, black-colored veins, less than 0.1 mm wide, are found traversing the matrix (Fig. 7), and numerous globules consisting of metallic nickel-iron, troilite and metal-troilite aggregate showing eutectoid intergrowth are interspersed in the devitrified silicate material composing veins (Fig. 8).

Most plagioclase grains in the vicinity of the veins possess deformational lamellae (Fig. 9), and several olivine grains intersected by the vein exhibit kink band in cross

polarized light. The microscopic feature of veins and the surrounding matrix both in transmitted and reflected light reveals that veining was produced by melting and mechanical deformation. The event gave a strong effect locally, and any textural transformation resulting from melting associated with stress is not observed in the matrix except at the veins and their vicinity. This fact suggests that veining was probably induced by the post-formational shock event (Begemann and Wlotska, 1969; Buseck, 1967; Fredriksson, *et al.*, 1963).

Olivine: Olivine, a dominant mineral constituent of Nagai, occurs in hypidiomorphic or xenomorphic shape. Optic axial angle is $2V = -85^\circ$ in average, and refractive indices are $\alpha = 1.678$ and $\gamma = 1.723$. Cell dimension is $a = 4.777 \text{ \AA}$, $b = 10.28 \text{ \AA}$ and $c = 6.017 \text{ \AA}$. The molar composition is estimated to be Fa_{25} by the optics. Twinned crystals possessing a twinning plane (011) are rarely found in the matrix.

Pyroxene: The main species of pyroxene is orthopyroxene. Optic axial angle is $2V = -72^\circ$ in average, and refractive indices are $\alpha = 1.682$ and $\gamma = 1.694$. Cell dimension is $a = 18.29 \text{ \AA}$, $b = 8.872 \text{ \AA}$ and $c = 5.203 \text{ \AA}$. The molar composition is Fs_{25} according to Kuno's (1954) curve. Clinopyroxene, probably augite, is present as a reaction rim, ranging from 0.01 mm to 0.05 mm in width, around orthopyroxene crystals, and also occurs as exsolution lamellae in orthopyroxene, sharing common (100) plane and [001] axis between clinopyroxene and the host orthopyroxene. The extinction angle ($c \wedge Z$) of the rim clinopyroxene is about 40° . Stillwater-typed exsolution lamellae are also frequently present in the clinopyroxene rim around orthopyroxene.

Plagioclase: Plagioclase, xenomorphic in shape, occurs as clear and transparent crystals which are poor in inclusions. Optic axial angle is $2V = -58^\circ$ in average, and refractive indices are $\alpha = 1.530$ and $\gamma = 1.541$. Optical data indicate a high-temperature plagioclase, and the molar composition is An_{13} according to optic axial angle data. Twinned crystals in albite, Carlsbad and Carlsbad-albite types are commonly found in Nagai.

Opaque minerals: Mineral species are nickel-iron, troilite and chromite. Nickel-iron grains consist usually of two separated phases, kamacite and taenite and troilite occurs as aggregates composed of differently oriented individuals. Opaque grains are generally xenomorphic in shape, but columnar and platy crystals of nickel-iron and troilite, about a few micrometers in size, which are surrounded by idiomorphic crystal planes are rarely present included in a few silicate grains.

IV. Conclusion

On the basis of the chemical composition, particularly from the results of the stepwise chemical fractional dissolution, it is concluded that the meteorite Nagai belongs to the L-group highly metamorphosed and recrystallized ordinary chondrite. This conclusion is further supported by the petrographical and mineralogical in-

vestigations. The well-developed chondritic texture of the chondrite, the presence of high-temperature plagioclase, poorly defined chondrules, and the uniform and homogeneous composition of olivine and orthopyroxene suggest that Nagai belongs to the petrologic class L6 of Van Schmus and Wood's classification (1967).

V. Acknowledgement

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VI. References

- BEGEMANN, F. and F. WLOTZKA, 1969. Shock induced thermal metamorphism and mechanical deformations in Ramsdorf chondrite. *Geochim. Cosmochim. Acta.* **33**: 1351-1370.
- BUSECK, P. R., 1967. The post-formational history of a hypersthene chondrite—Beenham. *Geochim. Cosmochim. Acta.* **31**: 1583-1587.
- DODD, R. T. Jr., W. R. Van Schmus and D. M. KOFFMAN, 1967. A survey of the unequilibrated ordinary chondrites. *Geochim. Cosmochim. Acta.* **31**: 921-951.
- FREDRIKSSON, K., P. S. DE CARLI and A. ARAMÄE, 1963. Shock-induced veins in chondrites. *In* Space Research III, W. Priestler Ed., pp. 974-983. North Holland.
- IKEDA, N., 1951. New colorimetric method for the determination of phosphate ion by sodium-thiosulphate. *Nippon Kagaku Zasshi (J. Chem. Soc. Japan, Pure Chem. Ed.)* **72**: 23-26 (in Japanese).
- IMAI, S., W. AIHARA, H. MIZUKAMI, Y. OMORI and T. MASUDA, 1978. Shimadzu inductively coupled plasma quantometer model ICPQ-100. *Shimadzu Review* **35**: 59-63. (in Japanese with English summary).
- KEIL, K. and K. FREDRIKSSON, 1964. The iron, magnesium and calcium distribution in coexisting olivines and rhombic pyroxenes of chondrites. *J. Geophys. Res.*, **69**: 3487-3515.
- KUNO, H., 1954. Study of orthopyroxenes from volcanic rocks. *Amer. Mineral.* **39**: 30-46.
- LEVI-DONATI, G. T., Masako SHIMA and G. P. SIGHINOLFI, 1976. Brazilian Meteorite: The Mafra, Santa Catarina State, chondrite. *Meteoritics* **11**: 29-41.
- MASON, B., 1963. Olivine compositions in chondrites. *Geochim. Cosmochim. Acta.* **27**: 1011-1024.
- , 1971. *Handbook of elemental abundances in meteorites.* Gordon & Breach Sci. Publ. New York, Paris, London.
- PRIOR, G. T., 1920. The classification of meteorites. *Mineral. Mag.* **19**: 51-63.
- SHIMA, Masako, 1974. The chemical composition of the stone meteorites Yamato (a), (b), (c) and (d) and Numakai. *Meteoritics* **9**: 123-135.
- , 1976. Mineral separation—Stepwise fractional chemical dissolution of minerals. *In* Shinjikken Kagaku Koza (New Experimental Procedure in Chemistry). *Cosmo-Geo-Chemistry*, pp. 248-257. Chemical Society of Japan Ed., Maruzen Publ. Co., Tokyo. (in Japanese).
- , 1979. The abundances of titanium, zirconium and hafnium in stony meteorites. *Geochim. Cosmochim. Acta.* **43**: No. 3 in Press.
- , K. P. JOCHUM, G. P. SIGHINOLFI and H. HINTENBERGER, 1974. The chemical composition of

major elements and heavy trace metals in chondrites Parambù and Marilia. *Meteoritics* **9**: 199-207.

———, A. OKADA, Y. OMORI, T. MASUDA and Y. TANAKA, 1978. Japanese Iron Meteorites: Chemical composition and microanalytical observation of Kuga, Shirahagi, Tanakami and Tendo. *This Journal*.

VAN SCHMUS, W. R. and J. A. WOOD, 1967. A chemical-petrologic classification for the chondritic meteorites. *Geochim. Cosmochim. Acta.* **31**: 747-765.

