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SIMS STUDIES ON ISOTOPIC ABUNDANCE ANOMALIES OF MAGNESIUM
IN PRIMITIVE METEORITES

BY

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PREFACE

The origin of the solar system has been interesting historically for a long period of time. For the study of the early solar system, extraterrestrial materials such as meteorites play an important role, because some of them contain informations about the primordial solar nebula.

Recently, isotopic studies on meteorites have brought fruitful results, among which, the most striking is on the idea of the multi-component solar nebula. First evidence was found by Clayton et al. through the study of the Allende meteorite which fell in 1969. They found the excess of ^{16}O in the meteorite, which was interpreted as the result of the injection of new component into the early solar nebula.

This paper describes results of study, which strongly supports the above mentioned idea, through the discovery of ^{24}Mg anomaly in the Allende and Yamato-74191 meteorites.

This paper consists of six sections and an appendix. Section one is a historical view and an introduction to the investigation of isotopic abundance anomaly of magnesium. Section two is a description of samples of meteorites and terrestrial minerals. Section three is a description of apparatus used in the present investigation, which are ion microprobe mass analyzers. In section four and section five, experimental procedures, results and discussion for magnesium isotopic abundance ratio measurements are described. Conclusion is stated in section six. In the appendix, computer programs developed by the author himself for a precise isotopic ratio measurement are listed.

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ABSTRACT

Isotopic abundance ratios of magnesium have been measured in the Allende (C3), Yamato-74191 (L3) and Yamato-75028 (H3) meteorites. Three Al-rich inclusions in Allende have been analyzed and excess ^{26}Mg due to the decay of now-extinct ^{26}Al has been found in the two of them. One of them has excess ^{26}Mg correlating with Al/Mg ratio. The other one has excess ^{26}Mg , but it is likely to diffuse out to the boundary layer by the heating after the specimen was formed. Al-correlated excess ^{26}Mg gives an estimate of $^{26}\text{Al}/^{27}\text{Al}$ ratio of $(2.8 \pm 2.2) \times 10^{-4}$ at the time when the minerals in the inclusion crystallized. This leads to the estimate of a time interval between the nucleosynthesis of ^{26}Al and the crystallization of the minerals of $(1.3^{+1.6}_{-0.6}) \times 10^6$ years.

Excess ^{24}Mg has been discovered for Al-poor and Mg-rich matrix areas of both Allende and Yamato-74191. The excess ^{24}Mg is suggestive of the nucleosynthesis of almost pure ^{24}Mg and its injection into the pre-existed solar nebula. And this almost pure ^{24}Mg is expected to be synthesized in an explosive carbon burning process on the occasion of a supernova explosion.

These isotopic abundance anomalies of ^{24}Mg and ^{26}Mg experimentally found in this work together with ^{16}O excess reported by Clayton et al. can be strong evidences to support the hypothesis that the primordial solar nebula was inhomogeneous and composed of at least two components.

1. HISTORICAL VIEW AND INTRODUCTION

Isotopic abundance anomalies in primitive meteorites play an important role in the understanding of the origin and the early history of the solar nebula.

Before 1960's it had been accepted that the primordial solar nebula was a single component and isotopically homogeneous.¹⁻³⁾ Meanwhile, isotopic abundance anomalies had long been searched in meteoritic and terrestrial samples in conjunction with the production of nuclei by the irradiation of energetic particles in the early history of the solar system.

In 1960, Reynolds⁴⁾ reported that the large excess of ^{129}Xe was ascribed to the radioactive decay of now extinct ^{129}I whose half life is 1.7×10^7 years. It was concluded that ^{129}I still existed at the time of the solidification of the chondrite. The time interval between the solidification and the nucleosynthesis was calculated to be $(3.5 \pm 0.6) \times 10^8$ years for Richardton assuming that the ratio of ^{129}I to ^{127}I at the termination of nucleosynthesis, $(^{129}\text{I}/^{127}\text{I})_0$, is equal to unity. This time interval has been called "formation interval".

Magnesium isotopic anomalies have been investigated especially for the excess of ^{26}Mg due to the decay of now extinct ^{26}Al (half life = 7.2×10^5 y), since ^{26}Al was thought to be possible heat source in melting meteorite parent bodies.⁵⁾ In spite of the search for ^{26}Mg anomaly,

neither excess nor depletion exceeding statistical errors have been observed until 1972.

In 1964, Shima⁷⁾ reported no variation of magnesium isotopic abundance for three terrestrial and one chondritic samples.

In 1970, Clarke et al.⁹⁾ reported that Bruderheim and Kohr Temiki chondrites had 4 to 6 % excess in ^{26}Mg . For the same samples, however, Schramm et al.¹⁰⁾ observed no anomaly of ^{26}Mg in 1970.

The Allende meteorite, which fell in Mexico in 1969,¹¹⁾ has brought an unexpected development in the investigation of the origin of the primordial solar nebula through isotopic anomaly studies. This meteorite has been classified into C3 carbonaceous chondrite, and much examined since its fall. It was confirmed mineralogically^{12,13)} and isotopically¹⁴⁻³²⁾ to contain primordial materials. The isotopic evidence of primitiveness firstly found was the anomaly of ^{16}O . The excess ^{16}O was observed for Ca-Al-rich white inclusions in Allende by Clayton et al.¹⁴⁾ They have described that the excess ^{16}O might be the result from the admixture of a component of almost pure ^{16}O . They thought that this component might represent interstellar material with a separate history of nucleosynthesis. Furthermore, ^{16}O anomalies have been found even in an ordinary chondrite ALHA-76004 of the type LL3 by Mayeda et al.¹⁵⁾

The attempts of searching for the excess ^{26}Mg which was a decay product from ^{26}Al have been successively carried

out for Allende by several workers. Gray and Compston¹⁶⁾ showed in 1974 that the excess ^{26}Mg was found to be 0.41 % in a chondrule with high Al/Mg ratio, and it was concluded to be due to the decay of ^{26}Al . In the same year, Lee and Papanastassiou¹⁷⁾ reported the ^{26}Mg anomalies, but the anomalies did not correlate with Al/Mg, and did not appear to be due to ^{26}Al decay. Lee et al.¹⁸⁾ reexamined in 1976 a Ca-Al-rich inclusion of Allende and observed the enrichment of ^{26}Mg up to 1.3 %. This excess has correlated with Al/Mg ratio and would be due to the in situ decay of ^{26}Al . From the extent of the excess and Al/Mg ratio, the initial ratio of $^{26}\text{Al}/^{27}\text{Al}$ at the time of the solidification of the inclusion was estimated to be about 6×10^{-5} . In 1979, Lee et al. further observed a larger excess of ^{26}Mg up to 9.7 % for anorthite mineral separates by using their direct loading technique.^{19,20)} Esat et al. also reported²¹⁾ highly fractionated Mg and the negative anomaly of ^{26}Mg , which has been designated FUN anomaly, for two Allende inclusions. Stegmann and Begemann reported in 1981²²⁾ the ^{26}Mg excess of 15 % correlating with Al concentration, and $^{26}\text{Al}/^{27}\text{Al}$ ratio at the time of solidification was estimated to be 5.9×10^{-5} .

The investigation mentioned above have been performed by using the method of thermal ionization mass spectrometry.

An ion microprobe mass spectrometric technique has been also used for the investigation of magnesium isotopic

abundance anomaly. Bradley et al.²³⁾ reported the excess ^{26}Mg up to 40 % in an anorthite grain picked out of the Allende meteorite with an ion microprobe. The excess has correlated with Al/Mg ratio. Hutcheon et al.²⁴⁾ reported that two anorthite specimens in Allende had ^{26}Mg excess of 7-18 %. Shimizu et al.²⁵⁾ also reported that ca. 13 % excess ^{26}Mg was observed in an anorthite inclusion and 16 % excess in a hibonite inclusion from Leoville C3 carbonaceous chondrite. In 1979, Hutcheon et al.²⁶⁾ showed that melilite and hibonite crystals from Allende had distinct ^{26}Mg excesses.

These data have been obtained by the use of ion microprobe techniques.

The discoveries of ^{16}O and ^{26}Mg anomalies in Ca-Al-rich inclusions of the Allende and Leoville carbonaceous chondrites have led to the search for more anomalies of other elements in order to clarify the special features of the Allende chondrite. In both 1977 and 1978, isotopic abundance anomalies of such elements as S,²⁷⁾ Ba and Nd,²⁸⁾ Ca,²⁹⁾ Sr,³⁰⁾ Sm³¹⁾ and Ag³²⁾ have been observed one after another in the Allende meteorite.

The excess of ^{26}Mg correlating with Al/Mg ratio has been concluded to be due to the in situ decay of ^{26}Al , and a view that ^{26}Al existed at the time when the minerals in the inclusion crystallized has been generally accepted. This ^{26}Al has been thought to be synthesized shortly before the crystallization because of the short half life

(7.2×10^5 y) of ^{26}Al . The time interval between the synthesis of these isotopes and the crystallization has been estimated to be at most a few million years.^{19,33)} This time interval is very much different from the ^{129}I - ^{129}Xe formation interval of ca. 2×10^8 years observed for Allende inclusions.^{34,35)} In order to explain this difference consistently, it has been considered that ^{26}Al has a separate origin from ^{129}I . This idea led to a hypothesis as follows:

Two separate nucleosynthetic events were considered. One of them would be around 2×10^8 years before the crystallization of minerals and the other, a few million years before. Both of the events have been considered to be supernova explosions, although these would have considerably different conditions. A supernova explosion which synthesized such r-process nuclei as ^{129}I would be accompanied by larger amount of neutron flux, and another one which synthesized ^{26}Al and ^{16}O would be a supernova whose predominant nuclear process was an explosive carbon burning and the latter did not give so much neutron flux as ^{129}I could be formed. Because, if the latter supernova formed enough ^{129}I , ^{129}I - ^{129}Xe and ^{26}Al - ^{26}Mg formation intervals should be essentially the same values.

If such a latter supernova as in the above mentioned idea had exploded, this would have synthesized ^{20}Ne , ^{23}Na , ^{24}Mg and ^{28}Si together with ^{16}O and ^{26}Al , and these nuclei would be injected into the primordial solar nebula from the

theoretical considerations by Arnett³⁷⁾ and Arnett and Truran.³⁸⁾

As a result of the formation and injection of these isotopes, their remnants may be kept in the primitive meteorites as in the case of ^{16}O and ^{26}Mg . The possibility led us to investigate not only ^{26}Mg isotopic abundance anomaly but also ^{24}Mg anomaly for primitive meteorites. We have investigated them with ion microprobe mass analyzers, because they are capable of an isotopic abundance measurement for a localized portion of the order of 100 μm size. This capability is essentially important for the study of such specimens as chondrites that are usually complex aggregates of various kinds of fine minerals.

In order to confirm the anomaly of ^{26}Mg and to obtain distribution of the anomaly on the sample surface, line analyses of magnesium isotopic ratios across three inclusions of the Allende meteorite have been carried out firstly. Secondly, a search for an excess ^{24}Mg , which has never been observed, was performed for many Al-poor and Mg-rich portions of matrix areas of a few primitive chondrites.

2. SAMPLES

Meteoritic samples used for the present investigation of isotopic abundance anomalies of ^{26}Mg and ^{24}Mg are listed in Table 2.1. Ten specimens of terrestrial mineral samples were also used and the list is in Table 2.2.

2.1 METEORITIC SAMPLES

Three meteoritic samples shown in Table 2.1 are chondrites. The classifications of those chondrites based on the categories proposed by Van Schmus and Wood³⁹⁾ are also listed in the second column of the table. C, L and H represent the classification based on Fe/Si ratio and the degree of oxidation. The degree of oxidation becomes lower in this order. C corresponds to carbonaceous chondrite, L, ordinary chondrite of low iron group, H, ordinary chondrite of high iron group. There are two other groups of E (enstatite chondrite) and LL (low iron and low metal ordinary chondrite), although chondrites of these two groups were not used in the present work. The number after the group name, 3 in this case, means the petrologic type proposed by Van Schmus and Wood.³⁹⁾ The petrologic type ranges from 1 to 6 (or recently 7) and represents the degree of metamorphism. The higher the number of petrologic type is, the more the metamorphism proceeds. A chondrite belonging to the petrologic type less than 3 is unequilibrated and is generally accepted to contain primordial

material, almost unaltered, although types 1 and 2 in H, L and LL groups have not been found.

2.1.1 Allende

A photomicrograph of a cut surface of the Allende (C3) carbonaceous chondrite is shown in Fig.2.1. Various kinds of inclusions, which are different in color, shape, size and texture, are embedded in black matrix. It has been reported that white inclusions are abundant in Al-rich refractory minerals, which are thought to have crystallized in the early stage of solidification from gaseous state^{42,43)} and to be primitive in case of the Allende chondrite.^{12,13)} Since white inclusions appear to have various thermal histories, judging from their profiles, the following three specimens with different shapes and textures were selected.

They are:

- (1) an amoeboid whitish inclusion,
- (2) a relatively small chondrule-like white inclusion surrounded by a ring-shaped boundary layer, and
- (3) a large round white inclusion.

Photomicrographs of these specimens are shown in Figs. 2.2, 2.3 and 2.4, respectively. The specimen 1 may have been heated and once melted from the amoeboid shape. The specimen 2 has a ring-shaped boundary layer which has a microscopically different texture from the central inclusion part. The specimen 3 is similar in size and texture to the specimen investigated by Phinney et al.,⁴⁴⁾ which contain

refractory-rich inclusions. These three specimens are abbreviated as AL0, AL1 and AL2, respectively, in this paper.

2.1.2 Yamato-74191

A photomicrograph of a cut surface of the Yamato-74191 (L3) chondrite is shown in Fig. 2.5. As is seen in the picture, most part of the surface is occupied by chondrules and there is a little matrix area which exists in the narrow portion among the chondrules.

This chondrule has been reported to be unequilibrated,^{41,45)} and to contain a large amount of trapped gases.⁴⁶⁾ Thus it is expected to contain primordial materials. This chondrite is abbreviated as Y-74191.

2.1.3 Yamato-75028

A photomicrograph of a cut surface of the Yamato-75028 chondrite is shown in Fig. 2.6. This chondrite itself is reported to be a breccia of H3 and L3 matter with H5 clasts.⁴¹⁾ A specimen of H3 part of this chondrite was distributed by the National Institute of Polar Research. Since the petrologic type is 3, this specimen is thought to be less metamorphosed. This is abbreviated Y-75028.

2.2 TERRESTRIAL SAMPLES

Terrestrial mineral samples listed in Table 2.2 are all silicates. A forsterite in dunite from Ehime Pref., Japan was used as a laboratory standard. Four olivine samples in

lherzolite or spinel lherzolite were used as sub-standards. Using the samples of a hornblende, a vesuvianite, and a cordierite together with the forsterite, calibration curves for Al/Mg and Mg/Si ratios were formed in secondary ion mass spectrometry. An anorthite and a feldspar samples were used for the examination of the interference of sodium in mass spectra. The details of the examination will be separately described in the later section.

For the investigation of ^{26}Mg isotopic abundance anomaly, three inclusions of Allende were analyzed. For the study of ^{24}Mg anomaly, matrix parts of the three chondrites were analyzed.

3. APPARATUS

Two ion microprobe mass analyzers were used for magnesium isotopic analyses. One of them is a home-made apparatus and the other is a Hitachi IMA 2A apparatus.

3.1 ION MICROPROBE MASS ANALYZER

An ion microprobe mass analyzer has several merits. They are:

- (1) Quantitative or semi quantitative elemental analysis of the localized portion of a solid sample is possible,
- (2) Isotopic analysis is possible,
- (3) No chemical treatment is necessary before the analysis except polishing and cleaning, and
- (4) The consumption of a sample is extremely low compared with that by a wet chemical method.

These merits are expected to be advantageous to the isotopic analysis of chondritic materials, which are heterogeneous and are aggregates of various kinds of fine minerals, because the selection of analyzed area is quite easy.

Schematic diagrams of a home-made and a Hitachi's apparatus are shown in Figs. 3.1 and 3.2, respectively. These two apparatus are essentially the same constitutions. The apparatus consists of an ion source for producing primary ions, accelerating and focusing electrodes for primary ion beam, a sample holder and its moving device, accelerating and focusing electrodes for secondary ions, a double

focusing mass spectrometer, and pumping systems.

3.2 PRIMARY ION GUN AND FOCUSING SYSTEM

The primary ion gun is of a hollow cathode type. An example of the ion source of the home-made apparatus is illustrated in Fig. 3.3. Typical operating conditions of the ion guns of both apparatus are listed in Table 3.1.

The accelerating and focusing system for primary ion beam consists of a drawing out and accelerating electrodes, two sets of Einzel lenses (objective and condenser lenses) and deflecting electrodes.

Typical working conditions for magnesium isotopic analysis are tabulated in Table 3.2.

3.3 SAMPLE HOLDER

A sample mounting system of the home-made apparatus is illustrated in Fig. 3.4. A tantalum plate was used as a holder. Samples were mounted on the holder, and the surfaces of the samples were covered with a sheet of tantalum with a slit of about 2 mm wide and 20 mm long as shown in the figure. The holder was movable perpendicular to the primary ion beam in Z direction shown in Fig. 3.4. The incident angle of the primary ion beam to the surface of the sample was 45° , and the drawing out direction of the secondary ions was also 45° to the surface normal.

A sample mounting system of the Hitachi IMA 2A is shown in Fig. 3.5. Samples were mounted on a cylindrical holder

and, were covered with a sheet of tantalum with apertures of 3 mm in diameter, through which samples were bombarded by primary ions. The incident direction of the primary ion beam is perpendicular to the sample surface, and the drawing out direction of the secondary ions was 45° to the surface normal.

3.4 COLD FINGER

For the precise isotopic ratio measurement, it is a problem that molecular ions may overlap the subject mass peak at the same mass number. Large part of molecular ions which may interfere the isotopic ratio measurement are originated from volatile gas adsorbed on the sample surface. In order to remove the volatile gas in the sample chamber and to decrease the interferences, a liquid nitrogen cold finger was put aside the sample holder in both the apparatus. A schematic diagram of the installation is shown in Fig. 3.6. The details of the examination of interferences will be described in a later section.

3.5 ELECTRON SPRAY

When a sample is an insulator, charge-up effect due to the positive ion bombardment causes the instability of secondary ion current. In order to avoid the charge-up, an electron spray was utilized. As the result, enough stable secondary ion current could be obtained. A chart showing the stability of secondary ion current is shown in Fig. 3.7. In the figure, the peak top of $^{24}\text{Mg}^+$ for a terrestrial

forsterite sample was recorded for about one hour. A short period irregular fluctuation is found to be less than 0.4 % in this case, although a slow variation with time is observed to be about 1.5 % for one hour. Since the slow variation can be corrected by a data acquisition procedure, there is no problem about the isotopic ratio measurement.

3.6 MASS SPECTROMETER AND PUMPING SYSTEM

The mass spectrometer is of a double focusing type. Radii and deflection angles of electric and magnetic sectors are listed in Table 3.3. Width of each slit under the usual working conditions are also shown in the table. Resolutions of these mass spectrometers used in the present work are shown in the same table.

Two pumping systems of the same constitutions are used in the home-made apparatus. It consists of an oil diffusion pump with a liquid nitrogen cold trap followed by a rotary pump.

In case of the Hitachi's apparatus, two systems of a turbomolecular pump with a liquid nitrogen cold trap followed by a rotary pump, an oil diffusion pump followed by a rotary pump, and an ion pump are used as shown in Fig. 3.6.

3.7 DETECTING SYSTEM

Mass-analyzed secondary ions were amplified by a secondary electron multiplier followed by an amplifier and a pen recorder.

The secondary ion intensity has been measured as a peak height of a mass spectrum.

3.8 COMPUTER CONTROLLING SYSTEM

A computer controlling system has been developed with a microcomputer for a precise isotopic ratio measurement. The control of mass scanings as well as the calculations of isotopic ratios and the statistical procedures can be performed. A block diagram of this controlling system is shown in Fig. 3.8.

A CPU used in this system is 8080AFC. Interfaces constructed are:

- (1) to operate a power supply of an electromagnet,
- (2) to put on and off a recorder,
- (3) to read the output DC voltage of an amplifier into computer as 3.5 digit BCD code,
- (4) to control an AD converter,
- (5) to switch sensitivities of detecting system by switching a feed back resistor of the amplifier, and
- (6) to read out the data from the computer to a digital printer.

These interfaces are shown in Figs. 3.9, 3.10 and 3.11.

A software of this controlling system has been also developed by the author. This program consists of three parts. These are:

- (1) a program of setting initial conditions which are re-

quired for the isotopic ratio measurement by the dialog between a computer and an operator through a CRT screen,

(2) a program of controlling the interfaces, and

(3) a program of calculating isotopic ratios and of a statistical procedure.

Programs (1) and (3) are written with BASIC and (2) is written with assembler. These programs are listed in Appendix.

The computer controlling system has been mainly used in Hitachi IMA 2A apparatus. The details of the working of this system will be stated in the later section.

4. EXPERIMENTAL

Isotopic abundance ratios of magnesium have been measured for the three chondritic samples comparing with those for terrestrial samples.

4.1 SAMPLE PREPARATION

Most samples shown in Table 2.1 and 2.2 were polished with emery papers and ultrasonically washed and cleaned in acetone. For some samples, freshly spalled surfaces were used without polishing and cleaning procedures.

Those samples were set on a holder as shown in Figs. 3.4 and 3.5.

4.2 EXAMINATION OF INTERFERENCES

Molecular and multiply-charged ions are usually formed as secondary ions by ion bombardment of a sample surface. Among these, ionic species overlapping a subject mass peak interfere the isotopic ratio measurement. Possible interfering ionic species at mass 24, 25 and 26 of subject ionic species of $^{24}\text{Mg}^+$, $^{25}\text{Mg}^+$ and $^{26}\text{Mg}^+$ are listed in the second column of Table 4.1.

These overlaps can be avoided in principle by making a mass resolution high enough to resolve these interfering species from the subject mass peak. Resolutions necessary to resolve each interfering ionic species are listed in the third column of Table 4.1. However, even if a high resolu-

tion is attained, the interfering peaks are in most cases masked by a tailing of the subject mass peak and they can be hardly detected, since the intensity of almost all interfering species are expected to be less than 1×10^{-3} of the subject peaks in the present work. Adding this, it is necessary to take an ion intensity as high as possible to minimize an error due to the statistical fluctuation of the intensity.

Taking this situation into account, we have carried out the measurements with low resolution mode and extensively examined the contributions of the interferences in the following manner.

4.2.1 $^{24}\text{MgH}^+$ and $^{25}\text{MgH}^+$

As described in the previous section, a liquid nitrogen cold finger was put aside the sample holder in order to decrease hydride ions. A remarkable effect of the cold finger was observed as shown in Table 4.2. The estimate was carried out according to eqs. (4.1) and (4.2) using the terrestrial forsterite and olivine samples.

$$\frac{I_{25}}{I_{24}} = \frac{A_{25}}{A_{24}} (1-\beta) + x \quad \dots\dots (4.1)$$

$$\frac{I_{26}}{I_{24}} = \frac{A_{26}}{A_{24}} (1-\beta)^2 + \frac{A_{25}}{A_{24}} x \quad \dots\dots (4.2)$$

where I_m : secondary ion intensity at mass number m ,

A_m : natural isotopic abundance of ^mMg ,

β : mass discrimination factor,

x: hydride ion formation ratio for Mg.

The values of x and β were obtained by solving these equations. The value of x is shown in Table 4.2, which is the maximum value of x's for the samples of a terrestrial forsterite and terrestrial olivines in four lherzolites. The value of β was 0.014 with a standard deviation of 0.002.

4.2.2 $^{23}\text{NaH}^+$ and $^{23}\text{NaH}_2^+$

The contribution of $^{23}\text{NaH}^+$ to the peak at mass 24 was estimated from mass spectra obtained for a terrestrial sodium-rich feldspar sample in granodiorite from Hyogo Pref., Japan and an anorthite sample from Hokkaido, Japan. Since the peak at mass 24 essentially consisted of two components, $^{24}\text{Mg}^+$ and $^{23}\text{NaH}^+$, in this case, $^{23}\text{NaH}^+$ was evaluated by subtracting the contribution of $^{24}\text{Mg}^+$. $^{24}\text{Mg}^+$ was estimated as follows. Neglecting x in eqs. (4.1) and (4.2), and after eq. (4.2) is divided by the square of eq. (4.1), we can get

$$\frac{I_{26} \cdot I_{24}}{(I_{25})^2} = \frac{A_{26} \cdot A_{24}}{(A_{25})^2} \quad \dots\dots (4.3)$$

The right term of eq. (4.3) is constant. Therefore, if I_{25} and I_{26} are substituted into eq. (4.3), then we can get I_{24} .

In almost all cases, no residue of the subtraction of

$^{24}\text{Mg}^+$ from the peak at mass 24 existed and the maximum value of $^{23}\text{NaH}^+ / ^{23}\text{Na}^+$ was found to be 5×10^{-5} . While $^{23}\text{Na}^+ / ^{24}\text{Mg}^+$ ratio was of the order of 1000 in case of the feldspar and anorthite samples, and the ratio was less than 0.05 in the usual case of chondrites, then the contribution of $^{23}\text{NaH}^+$ to $^{24}\text{Mg}^+$ was estimated to be less than 2.5×10^{-6} .

As for the contribution of $^{23}\text{NaH}_2^+$, even if the peak at mass 25 was all due to $^{23}\text{NaH}_2^+$ for the sodium-rich feldspar, the ratio of the intensity at mass 25 to that at mass 23 was less than 2.5×10^{-4} , although most part of the peak at mass 25 is observed to be originated from an impurity magnesium in the feldspar judging from the peak at mass 26 ($^{26}\text{Mg}^+$). Thus, $^{23}\text{NaH}_2^+ / ^{25}\text{Mg}^+$ in case of chondrite analysis was estimated to be much less than 1.5×10^{-4} .

4.2.3 $^{12}\text{C}_2^+$, $^{12}\text{C}^{13}\text{C}^+$, $^{13}\text{C}_2^+$, $^{12}\text{C}^{14}\text{N}^+$, $^{12}\text{C}^{13}\text{CH}^+$ and $^{12}\text{C}_2\text{H}_2^+$

The contributions of these ionic species were estimated from mass spectra obtained for five carbon-rich samples. The samples used are all terrestrial and are listed in the first column of Table 4.3. The contribution of $^{12}\text{C}_2\text{H}^+$ will be described in section 4.2.4.

Secondary ion intensities at masses 12, 12.5, 24, 25 and 26 were used for the estimate. The intensity of $^{12}\text{C}^+$ was evaluated by subtracting $^{24}\text{Mg}^{2+}$ estimated based on the intensity at mass 12.5 ($^{25}\text{Mg}^{2+}$) from the intensity at mass 12.

The diatomic ion formation ratio for carbon isotopes is evaluated as

$$^{12}\text{C}_2^+ : ^{12}\text{C}^{13}\text{C}^+ : ^{13}\text{C}_2^+ = 100:2.2:0.013 \quad \dots (4.4)$$

from the arithmetic combination of the isotopic abundances of carbon. Assuming that

$$^{12}\text{C}_2^+ : (^{13}\text{C}_2^+ + ^{12}\text{C}^{14}\text{N}^+ + ^{12}\text{C}^{13}\text{CH}^+ + ^{12}\text{C}_2\text{H}_2^+) = 100:y,$$

the following equations can be set up by neglecting the contributions of magnesium hydride ions as described in 4.2.1 and 4.2.2.

$$\frac{I_{25-2.2k}}{I_{24-100k}} = \frac{A_{25}}{A_{24}} (1-\beta) \quad \dots (4.5)$$

$$\frac{I_{26-yk}}{I_{24-100k}} = \frac{A_{26}}{A_{24}} (1-\beta)^2 \quad \dots (4.6)$$

where I_m : secondary ion intensity at mass m ,

A_m : natural isotopic abundance of ^mMg ,

β : mass discrimination factor,

k : proportional constant.

If we substitute the mass discrimination factor of 0.014 into β , which was obtained for the terrestrial forsterite sample, then the simultaneous equations (4.5) and (4.6) for the variables k and y can be solved. Using the obtained values of k and y , intensities of $^{12}\text{C}_2^+$, $^{12}\text{C}^{13}\text{C}^+$ and $(^{13}\text{C}_2^+ + ^{12}\text{C}^{14}\text{N}^+ + ^{12}\text{C}^{13}\text{CH}^+ + ^{12}\text{C}_2\text{H}_2^+)$ have been able to be evaluated. From the results, the molecular ion formation ratio of these ionic species to $^{12}\text{C}^+$ have been obtained as shown in the second and third columns in Table 4.3.

In cases of magnesium isotopic measurements for chondrite samples analyzed so far, the upper limit of the secondary ion intensity ratio of $^{12}\text{C}^+ / ^{24}\text{Mg}^+$ was 7×10^{-4} . From this value and the maximum value among the ratios listed in Table 4.3, the maximum contributions of $^{12}\text{C}^+$, $^{12}\text{C}^{13}\text{C}^+$ and $(^{13}\text{C}_2^+ + ^{12}\text{C}^{14}\text{N}^+ + ^{12}\text{C}^{13}\text{CH}^+ + ^{12}\text{C}_2\text{H}_2^+)$ to the respective subject peak were estimated as shown in Table 4.1.

4.2.4 $^{12}\text{C}_2\text{H}^+$

From the mass spectra obtained for the carbon-rich samples listed in Table 4.3, even if the peak appearing at mass 25 were all $^{12}\text{C}_2\text{H}^+$, the ratio of this intensity to $^{12}\text{C}^+$ was estimated to be less than 2×10^{-3} . Thus $^{12}\text{C}_2\text{H}^+ / ^{25}\text{Mg}^+$ was estimated to be less than 1×10^{-5} for the chondrite analysis.

4.2.5 $^{48}\text{Ca}^{2+}$

Calcium has six stable isotopes of masses 40, 42, 43, 44, 46 and 48. The doubly-charged ions of $^{40}\text{Ca}^{2+}$ appears at mass 20. The intensity ratios of $^{40}\text{Ca}^{2+} / ^{40}\text{Ca}^+$ was observed to be less than 0.015 for the chondrite samples used. Since $^{48}\text{Ca}^{2+} / ^{48}\text{Ca}^+$ would be equal to $^{40}\text{Ca}^{2+} / ^{40}\text{Ca}^+$, the contribution of $^{48}\text{Ca}^{2+}$ to $^{24}\text{Mg}^+$ was calculated to be less than 2×10^{-5} from the intensities at masses 24 and 48.

4.2.6 $^{48}\text{Ti}^{2+}$ and $^{50}\text{Ti}^{2+}$

Titanium has five stable isotopes of masses 46, 47, 48,

49 and 50. Doubly-charged ions of $^{47}\text{Ti}^{2+}$ and $^{49}\text{Ti}^{2+}$ appear at mass numbers 23.5 and 24.5, respectively. In cases of the analyzed chondrites, no peaks exceeding the noise levels at those mass numbers were observed when the sensitivities at mass numbers 23.5 and 24.5 were 10^3 times higher than that at mass 24. Therefore, the noise levels were taken as maximum intensities of $^{47}\text{Ti}^{2+}$ and $^{49}\text{Ti}^{2+}$. Thus, $^{48}\text{Ti}^{2+}/^{24}\text{Mg}^+$ and $^{50}\text{Ti}^{2+}/^{25}\text{Mg}^+$ were estimated to be less than 7×10^{-5} and 4×10^{-5} , respectively.

4.2.7 $^{50}\text{Cr}^{2+}$ and $^{52}\text{Cr}^{2+}$

Chromium has four stable isotopes of masses 50, 52, 53 and 54. Doubly-charged ions of $^{53}\text{Cr}^{2+}$ appears at mass 26.5. No peak could be detected even with 10^3 times higher sensitivity than that at mass 24. From the noise level at mass 26.5, $^{50}\text{Cr}^{2+}/^{25}\text{Mg}^+$ and $^{52}\text{Cr}^{2+}/^{26}\text{Mg}^+$ were estimated to be less than 1×10^{-6} and 5×10^{-4} , respectively.

4.2.8 Summary

The estimate of the contribution of doubly-charged and molecular ions was done for the Hitachi IMA 2A apparatus as mentioned above. For the home-made apparatus, the same way of the estimate was used. The maximum contribution of these interfering ionic species to the subject peak are summarized in the fourth and fifth columns in Table 4.1. From the results, the extent of the interferences is found to be less than 1 permil as a whole for each subject peak of $^{24}\text{Mg}^+$, $^{25}\text{Mg}^+$ and $^{26}\text{Mg}^+$.

4.3 CALIBRATION CURVES FOR Al/Mg AND Mg/Si CONCENTRATION RATIOS

In order to estimate the concentration ratios of Al/Mg and Mg/Si from the secondary ion intensity ratios of $^{27}\text{Al}^+ / ^{24}\text{Mg}^+$ and $^{24}\text{Mg}^+ / ^{28}\text{Si}^+$, calibration curves for these elemental concentration ratios have been prepared. Samples used were terrestrial forsterite, hornblende, vesuvianite and cordierite. These four minerals are all silicates and their localities are listed in Table 2.2. The concentrations of Mg, Al and Si determined by the analysis of atomic absorption spectrometry were tabulated in Table 4.4 with their chemical formulae.

Figures 4.1 and 4.2 are the graphs of secondary ion intensity ratios versus concentration ratios. Plotted points fall well on a straight line with the slope of unity. These lines can be used to determine concentration ratios of Al/Mg and Mg/Si.

Al/Mg ratio can be a measure for aluminum concentration and Mg/Si ratio can be used in order to select a forsterite-rich portion.

4.4 MAGNESIUM ISOTOPIC ANALYSIS

Magnesium isotopic analyses have been carried out for the chondritic samples described in section 2. Mass scan-nings were repeated 40 to 90 times for one probed portion over the mass range from mass 24 to 26. A peak at mass 27 has also been taken as a measure of Al concentration at

least once in a run. Examples of mass spectra are shown in Figs. 4.3 and 4.4. Figure 4.3 shows a mass spectrum obtained for the white inclusion of AL2 with the home-made apparatus, and Fig. 4.4, for a portion of the matrix of Y-74191 with the Hitachi IMA 2A apparatus.

Because the contributions of the interfering peaks to mass 24, 25 and 26 were found to be less than 1 permil as described in section 4.2, and to be negligibly small, raw secondary ion intensity ratios of magnesium were calculated from the mass spectra.

4.4.1 Definition of the deviation of an isotopic ratio

The deviation of an isotopic ratio of ${}^m\text{Mg}/{}^n\text{Mg}$ for a sample from the ratio for a reference is usually defined as eq.(4.7).

$$\Delta_{m/n} = \left(\frac{({}^m\text{Mg}/{}^n\text{Mg})_s}{({}^m\text{Mg}/{}^n\text{Mg})_r} - 1 \right) \times 1000 \quad \dots\dots (4.7)$$

where m and n are mass numbers of magnesium isotopes, and subscripts s and r represent "sample" and "reference", respectively.

The terrestrial forsterite (FO) in dunite from Ehime Pref., Japan was used to obtain the reference values of the isotopic ratios.

4.4.2 Automation of isotopic measurement by a microcomputer

The isotopic ratio has been measured automatically by

using a microcomputer. The hardware of the measuring system has been already described in section 3.6. Therefore, the details of the working of the system will be stated here mainly as to the software.

The working contains:

(1) Switching the scan speed, slow or fast, at the peak top and bottom, independently. In the usual case, the top was scanned slowly and the bottom, fastly.

(2) Reading the data into the memory by ten-point running mean method.

(3) Evaluating the peak height by the calculation of a top value minus a bottom value.

(4) Correcting the slow intensity variation with time during the scannings.

(5) Calculating the peak height ratios and Δ values. The formula of the calculation was preset in the computer.

(6) Taking statistical procedures that are the calculation of a mean, a standard deviation (σ) and a standard deviation of a mean (σ_m), the exclusion of data exceeding $\pm 2\sigma$, the recalculation of a mean and errors, and the iteration of these procedures.

(7) Printing the data with a digital printer.

A flow chart of the procedure is shown in Fig. 4.5.

4.4.3 Analysis of terrestrial standard

In both the apparatus, the laboratory standard sample of terrestrial forsterite (FO) has been intermittently ana-

lyzed over about one year in order to check the reproducibility for magnesium isotopic ratio measurements in both the apparatus.

$\Delta_{25/24}$ and $\Delta_{26/24}$ values obtained with the home-made apparatus are plotted in Fig. 4.6 in the chronological order. The weighted means of all the data of $^{25}\text{Mg}/^{24}\text{Mg}$ were evaluated to be 0.12495 for $m=25$ and 0.13666 for $m=26$, and these values were used as reference values in eq. (4.7). The reproducibilities were evaluated to be $\pm 3.5\%$ for $\Delta_{25/24}$ and $\pm 5.5\%$ for $\Delta_{26/24}$ as $\pm 2\sigma$. These values are shown as error bars for the closed circles which correspond to the laboratory standard values mentioned above as weighted means.

In Fig. 4.7, are replotted these Δ values as a three isotope plot. A straight line with the slope of 1/2 in the figure corresponds to the normal mass fractionation line. The plotted points are along the straight line. This means that the scattering of the data was originated by the normal mass fractionation, and that these data can be corrected by the normalization for the mass fractionation.

In Fig. 4.7, two points symbolized by a closed triangle and a closed square are also plotted. These two points correspond to the absolute isotopic abundance ratios of magnesium reported by Catanzaro et al.⁴⁷⁾ (CMGS 66) and Schramm et al.⁹⁾ (STW 70).

The weighted mean of our data (closed circle) deviates from these two data points by about 13 % in $\Delta_{25/24}$. This

deviation can be thought to be mainly due to a mass discrimination effects. It is not necessary to correct the mass discrimination in comparing the isotopic abundance ratios obtained by one apparatus under the same experimental conditions with each other.

In order to clarify the anomaly of $\Delta_{26/24}$, it had better be corrected for the normal mass fractionation. For the correction, the isotopic ratios for the laboratory standard of the terrestrial forsterite (FO) was used as a reference values. The following equations were used for the calculation of a normalized isotopic abundance ratio of $(^{26}\text{Mg}/^{24}\text{Mg})_n$ and its deviation, $\delta_{26/24}$, from the reference value:

$$\left(\frac{^{26}\text{Mg}}{^{24}\text{Mg}}\right)_n = \frac{(^{26}\text{Mg}/^{24}\text{Mg})_s}{(1-\alpha)^2} \quad \dots (4.8)$$

$$1-\alpha = \frac{(^{25}\text{Mg}/^{24}\text{Mg})_s}{(^{25}\text{Mg}/^{24}\text{Mg})_r} = 1 + \frac{\Delta_{25/24}}{1000} \quad \dots (4.9)$$

$$\delta_{26/24} = \left[\frac{(^{26}\text{Mg}/^{24}\text{Mg})_n}{(^{26}\text{Mg}/^{24}\text{Mg})_r} - 1 \right] \times 1000 \quad \dots (4.10)$$

where α is the mass fractionation factor. A subscript of n denotes "after normalization", s, "sample", and r, "reference".

The $\delta_{26/24}$ values for the same data as those plotted in Fig. 4.6 are plotted in Fig. 4.8 also in the chronological order. The reproducibility for $\delta_{26/24}$ was found to be $\pm 4.5\%$ as $\pm 2\sigma$.

$\Delta_{24/25}$ and $\Delta_{26/25}$ values obtained by the Hitachi IMA 2A apparatus are plotted in Fig. 4.9 in the chronological order. These data were also obtained by the analysis of the FO sample. From these data, weighted means of $^{24}\text{Mg}/^{25}\text{Mg}$ values were calculated to be 8.0178 for $m=24$ and 1.0872 for $m=26$. Error bars for the closed circles in the figure represent reproducibilities as $\pm 2\sigma$. Those are $\pm 2.8\%$ for both the Δ values as shown in the figure.

The absolute isotopic abundance ratios of $^{24}\text{Mg}/^{25}\text{Mg}$ and $^{26}\text{Mg}/^{25}\text{Mg}$ are 7.89702 and 1.10404⁹⁾, respectively. The weighted means of the ratios obtained with the Hitachi apparatus also deviate from the absolute ones. This is again caused mainly by mass discrimination effects. Although the correction for the mass discrimination is possible, it is not necessary to correct in the case that only the deviations of the isotopic ratios are compared with each other. It is enough to take a correction for the normal mass fractionation in the same sense as stated above.

Therefore, the values shown in Table 4.5 obtained for the sample of the FO were used as the reference values.

Moreover, four other terrestrial olivines, SL45, SL46, MM and OK listed in Table 2.2 were analyzed as sub-standards with the Hitachi IMA 2A apparatus. The data normalized to the reference values for FO are plotted in Fig. 4.10. In the figure, a normal mass fractionation line whose slope is -1 through the origin is illustrated.

The plotted points for those terrestrial sub-standards

fall well along the normal mass fractionation line. These points deviate by 3 - 6 % from the reference value. An explanation for the deviation guessed is as follows. The FO sample was taken from a rock of dunite and the four olivine samples were picked out of rocks of lherzolite or spinel lherzolite, and the localities of the rocks are all different. Because the differences of the types and the localities of rocks, the extent of mass fractionation may differ from one to another.

If the data for four olivine samples are corrected for the normal mass fractionation, the corrected values become consistent with that of FO standard within errors.

4.4.4 Search for ^{26}Mg anomaly

In order to search for ^{26}Mg anomaly, three specimens in the Allende carbonaceous chondrite, AL0, AL1 and AL2, have been used. There are descriptions about these three specimens. They all contain white or whitish inclusions which are expected to be composed of Al-rich refractory minerals such as hibonite, melilite and spinel.

Sketches of these inclusions are illustrated in Figs. 4.11, 4.12 and 4.13. The probed line for the magnesium isotopic analysis is shown in each figure. Photomicrographs of these inclusions have already been shown in section 2.

$\Delta_{\text{m}/24}$ values were calculated according to eq. (4.7) from raw secondary ion intensities. $^{27}\text{Al}^+ / ^{24}\text{Mg}^+$ ratio was also calculated. Furthermore, in order to obtain the extent

of ^{26}Mg anomaly, the normal mass fractionation was corrected by using the isotopic ratio for FO sample as the reference value. This correction was done by eqs. (4.8), (4.9) and (4.10).

4.4.5 Search for ^{24}Mg anomaly

In order to search for ^{24}Mg anomaly, the specimens of AL, Y-74191 and Y-75028 have been used. Al-poor and Mg-rich portions of matrix areas were analyzed in order to avoid the contribution from excess ^{26}Mg due to the decay of ^{26}Al . About 100 portions of matrix area were randomly analyzed for each specimen. An example of the probed portion is shown on the photograph in Fig. 2.1 by a red cross mark. Not only mass peaks at mass 24, 25 and 26 but also those at masses 27 and 28 were recorded at every probed portion..

$\Delta_{m/25}$ values were calculated by eq. (4.7). $^{27}\text{Al}^+ / ^{24}\text{Mg}^+$ and $^{24}\text{Mg}^+ / ^{28}\text{Si}^+$ ratios were also calculated. From Al/Mg ratio, portions with low aluminum concentrations could be found, and from Mg/Si ratio, forsterite-rich portions could be easily selected.

5. RESULTS AND DISCUSSION

5.1 ^{26}Mg ANOMALY

$\delta_{26/24}$ values obtained for the inclusions in the three specimens (AL0, AL1 and AL2) of the Allende chondrite are plotted in the chronological order in Figs. 5.1, 5.2, 5.3 and 5.4. The definition of the $\delta_{26/24}$ has appeared in the previous section and the $^{26}\text{Mg}/^{24}\text{Mg}$ ratio for FO was taken as the reference value.

The data in Fig. 5.3 correspond to the line analysis along AA' of the AL2 specimen in Fig. 4.13, and the data in Fig. 5.4, along BB' in the same specimen. In the lower part of Fig. 5.3, the data obtained for a terrestrial hornblende (HO) and vesuvianite (VE) are also shown.

These data for Allende are replotted in Figs. 5.5, 5.6, 5.7 and 5.8 as a function of the probed portion. $^{27}\text{Al}^+ / ^{24}\text{Mg}^+$ ratio for each probed portion is also shown as a measure of aluminum to magnesium concentration ratio.

5.1.1 Amoeboid whitish inclusion in AL0

As shown in Fig. 5.5, Al/Mg ratio is high in the area of this whitish inclusion. the anomaly of ^{26}Mg is not clear, although the mean value of $\delta_{26/24}$'s seems to be positive a little. The variation of $\delta_{26/24}$ does not correlate with that of Al/Mg ratio. Therefore, it was concluded that the excess ^{26}Mg could be hardly detected for this inclusion.

5.1.2 Chondrule-like white inclusion in AL1

The excess ^{26}Mg was observed rather in the ring-shaped boundary layer than in the white inclusion. There seems to be an anticorrelation between variations of $\delta_{26/24}$ and Al/Mg ratio except the data for the matrix area. From the variation of $\delta_{26/24}$ shown in Fig. 5.6, it may be considered that the white inclusion particle with ^{26}Al was firstly formed and after the formation of the particle, it was surrounded by the boundary layer. Then it was heated to enough high temperature not to melt but to recrystallize. During the recrystallization process, ^{26}Mg might be segregated toward the ring-shaped boundary layer. As the result of this segregation, excess ^{26}Mg was thought to be concentrated in the boundary layer.

5.1.3 Large white inclusion in AL2

This large white inclusion was analyzed along two different probed lines approximately perpendicular with each other. In both the analyses, excess ^{26}Mg more than 20 % was observed, and the data showed the correlation with the Al/Mg ratio as shown in Figs. 5.7 and 5.8.

Figures 5.9 and 5.10 are three isotope plots of magnesium for the white inclusion along the probed lines of AA' and BB', respectively. The data for the matrix area are omitted in the figures. The ordinate represents $\Delta_{25/24}$, and the abscissa, $\Delta_{26/24}$, calculated according to eq. (4.7). In each figure, a normal mass fractionation line with the slope

of 1/2 is illustrated through the origin which was determined from the data for the laboratory standard (FO). All the data are found to clearly fall in the right hand side of the normal mass fractionation line.

The deviation in the direction of the abscissa between each data point and the normal mass fractionation line corresponds to the $\delta_{26/24}$ value corrected for the normal mass fractionation. The deviation to the right side clearly shows the positive anomaly of ^{26}Mg .

In order to quantify a correlation between the excess ^{26}Mg and Al/Mg ratio, these data are replotted as shown in Fig. 5.11. The upper graph of $\Delta_{26/24}$ versus Al/Mg clearly shows the correlation that $\Delta_{26/24}$ increases with the increase of Al/Mg ratio. On the contrary, $\Delta_{25/24}$ shows no systematic variation in a whole range of Al/Mg in this figure.

5.2 Al-CORRELATED EXCESS ^{26}Mg

the Al-correlated excess ^{26}Mg , which was observed in the large white inclusion in AL2 specimen of the Allende chondrite as shown in Fig. 5.11, can be thought to be originated from the in situ decay of ^{26}Al . Using the plot in Fig. 5.11, $^{26}\text{Al}/^{27}\text{Al}$ ratio at the time of the crystallization of Al-containing minerals from the remnant materials of the exploded supernova as stated in sections 1 and 2 can be estimated.

5.2.1 ^{26}Al at the time of primordial mineral crystallization

If the excess ^{26}Mg due to the in situ decay of ^{26}Al is written as $^{26}\text{Mg}^*$, ^{26}Mg existed in a specimen can be expressed in the following equation.

$$(^{26}\text{Mg})_p = (^{26}\text{Mg})_o + ^{26}\text{Mg}^* \quad \dots\dots (5.1)$$

where subscript p represents "present", and o, "original" which means the initial magnesium originally existed.

When eq. (5.1) is divided by $(^{24}\text{Mg})_o$,

$$\frac{(^{26}\text{Mg})_p}{(^{24}\text{Mg})_o} = \frac{(^{26}\text{Mg})_o}{(^{24}\text{Mg})_o} + \frac{^{26}\text{Mg}^*}{(^{24}\text{Mg})_o} \quad \dots\dots (5.2)$$

Since $(^{24}\text{Mg})_o$ would be equal to $(^{24}\text{Mg})_p$ in this case, eq. (5.2) can be rewritten as

$$\left(\frac{^{26}\text{Mg}}{^{24}\text{Mg}}\right)_p = \left(\frac{^{26}\text{Mg}}{^{24}\text{Mg}}\right)_o + \left(\frac{^{26}\text{Mg}^*}{^{24}\text{Mg}}\right)_p \quad \dots\dots (5.3)$$

From eq. (5.3),

$$\frac{(^{26}\text{Mg}^*/^{24}\text{Mg})_p}{(^{26}\text{Mg}/^{24}\text{Mg})_o} = \frac{(^{26}\text{Mg}/^{24}\text{Mg})_p}{(^{26}\text{Mg}/^{24}\text{Mg})_o} - 1 \quad \dots\dots (5.4)$$

is derived. The right side of eq. (5.4) is the same form as the definition of $\Delta_{26/24}$ and if this is put by $(\Delta_{26/24})_p$, then

$$\frac{(^{26}\text{Mg}^*/^{24}\text{Mg})_p}{(^{26}\text{Mg}/^{24}\text{Mg})_o} = \frac{(\Delta_{26/24})_p}{1000} \equiv (\Delta_{26/24}')_p \quad \dots\dots (5.5)$$

Since $^{26}\text{Mg}^*$ was originally ^{26}Al at the time of the primor-

dial material crystallization,

$$^{26}\text{Mg}^* = (^{26}\text{Al})_c \quad \dots (5.6)$$

where subscript c means "crystallization".

Using eqs. (5.5) and (5.6),

$$\begin{aligned} (\Delta_{26/24}')_p &= \frac{(^{26}\text{Al})_c / (^{24}\text{Mg})_p}{(^{26}\text{Mg}/^{24}\text{Mg})_o} \\ &= \frac{(^{26}\text{Al}/^{27}\text{Al})_c ((^{27}\text{Al})_c / (^{24}\text{Mg})_p)}{(^{26}\text{Mg}/^{24}\text{Mg})_o} \end{aligned}$$

Since ^{27}Al is constant, so $(^{27}\text{Al})_c = (^{27}\text{Al})_p$, then

$$(\Delta_{26/24}')_p = \frac{(^{26}\text{Al}/^{27}\text{Al})_c}{(^{26}\text{Mg}/^{24}\text{Mg})_o} \times \left(\frac{^{27}\text{Al}}{^{24}\text{Mg}} \right)_p \quad \dots (5.7)$$

As $(^{27}\text{Al}/^{24}\text{Mg})_p$ can be expressed as $(^{27}\text{Al}/^{24}\text{Mg})_s$, where subscript s means "specimen". Therefore eq. (5.7) becomes:

$$(\Delta_{26/24}')_p = \frac{(^{26}\text{Al}/^{27}\text{Al})_c}{(^{26}\text{Mg}/^{24}\text{Mg})_o} \times \left(\frac{^{27}\text{Al}}{^{24}\text{Mg}} \right)_s \quad \dots (5.8)$$

By the calibration curve in Fig. 4.1, the concentration ratio of $(^{27}\text{Al}/^{24}\text{Mg})_s$ can be related to the secondary ion intensity ratio as

$$(^{27}\text{Al}/^{24}\text{Mg})_s = (^{27}\text{Al}^+/^{24}\text{Mg}^+)_s \times 0.71 \quad \dots (5.9)$$

When eq. (5.9) is substituted into eq. (5.8), we can get

$$(\Delta_{26/24}')_p = \frac{(^{26}\text{Al}/^{27}\text{Al})_c}{(^{26}\text{Mg}/^{24}\text{Mg})_o} \times \left(\frac{^{27}\text{Al}^+}{^{24}\text{Mg}^+} \right)_s \times 0.71 \quad (5.10)$$

thus, using eqs. (5.5) and (5.10),

$$(\Delta_{26/24})_p = k \times \frac{({}^{26}\text{Al}/{}^{27}\text{Al})_c}{({}^{26}\text{Mg}/{}^{24}\text{Mg})_o} \times \left(\frac{{}^{27}\text{Al}^+}{{}^{24}\text{Mg}^+}\right)_s \dots (5.11)$$

where $k = 7.1 \times 10^2$.

In this equation, $k \times ({}^{26}\text{Al}/{}^{27}\text{Al})_c / ({}^{26}\text{Mg}/{}^{24}\text{Mg})_o$ is the slope of the straight line in $\Delta_{26/24}$ vs $({}^{27}\text{Al}^+ / {}^{24}\text{Mg}^+)_s$ plot of Fig. 5.11. If we put this slope g , then

$$g = k \times \frac{({}^{26}\text{Al}/{}^{27}\text{Al})_c}{({}^{26}\text{Mg}/{}^{24}\text{Mg})_o} \dots (5.12)$$

and we can get an equation

$$\left(\frac{{}^{26}\text{Al}}{{}^{27}\text{Al}}\right)_c = \frac{g}{k} \times \left(\frac{{}^{26}\text{Mg}}{{}^{24}\text{Mg}}\right)_o \dots (5.13)$$

where $({}^{26}\text{Mg}/{}^{24}\text{Mg})_o$ means the absolute isotopic abundance ratio of magnesium, and this $({}^{26}\text{Mg}/{}^{24}\text{Mg})_o$ can be replaced by the ratio reported by Catanzaro et al.⁴⁷⁾ Then

$$\left(\frac{{}^{26}\text{Al}}{{}^{27}\text{Al}}\right)_c = \frac{0.13932}{7.1 \times 10^2} g \dots (5.14)$$

can be given.

From Fig. 5.11, the slope of $\Delta_{26/24}$ vs ${}^{27}\text{Al}^+ / {}^{24}\text{Mg}^+$ plot, g , is evaluated to be 1.4 ± 1.1 by the least square fit. The obtained straight line by the least square fit is illustrated in the upper graph of Fig. 5.11 with a solid line. In the figure, the upper and lower limits obtained from the error of standard deviation are also illustrated with dashed lines.

The value of g and eq. (5.14) give the result that

$$\left(\frac{{}^{26}\text{Al}}{{}^{27}\text{Al}}\right)_c = (2.8 \pm 2.2) \times 10^{-4} \quad \dots (5.15)$$

Lee et al.¹⁸⁾ reported that $({}^{26}\text{Al}/{}^{27}\text{Al})_c$ is about 0.5×10^{-4} . This value is a little different from the present result, but in consideration of the error limit of our data, these two are barely consistent with each other.

If our 2.8×10^{-4} is valid for the white inclusion in AL2, it is about 4.5 times higher than that by Lee et al. A possible explanation for this discrepancy is that the minerals investigated at present had crystallized about 1.6×10^6 years prior to the crystallization of the minerals for which Lee et al. have obtained their data.

5.2.2 Chronology from the viewpoint of ${}^{26}\text{Al}$ - ${}^{26}\text{Mg}$

The result stated in section 5.2.1 shows that ${}^{26}\text{Al}$ existed at the time of primordial mineral crystallization. Since the half-life of ${}^{26}\text{Al}$ is 0.72×10^6 years, it can be considered that the time interval between the nucleosynthesis of ${}^{26}\text{Al}$ and the crystallization of Al-rich minerals was comparable with the half life. This time interval can be estimated by the same method as the case of ${}^{129}\text{I}$ - ${}^{129}\text{Xe}$ chronology.

If we denote ${}^{26}\text{Al}/{}^{27}\text{Al}$ ratio at the time of nucleosynthesis as $({}^{26}\text{Al}/{}^{27}\text{Al})_0$, then

$$\left(\frac{{}^{26}\text{Al}}{{}^{27}\text{Al}}\right)_c = \left(\frac{{}^{26}\text{Al}}{{}^{27}\text{Al}}\right)_o \exp(-\lambda \Delta t) \quad \dots\dots (5.16)$$

where λ : decay constant of ${}^{26}\text{Al}$

Δt : time interval between the nucleosynthesis and the crystallization.

From eq. (5.16), we can get the following equation:

$$\Delta t = \frac{1}{\lambda} \left[\ln\left(\frac{{}^{26}\text{Al}}{{}^{27}\text{Al}}\right)_o - \ln\left(\frac{{}^{26}\text{Al}}{{}^{27}\text{Al}}\right)_c \right] \quad \dots\dots (5.17)$$

$({}^{26}\text{Al}/{}^{27}\text{Al})_o$ ratio has been reported by many workers based on theoretical considerations. The reported ratios are listed in Table 5.1 in the order of the year of publication.

Four recent reports give the ratios around 1×10^{-3} . These theoretical data have been obtained by the consideration of an explosive burning nucleosynthesis with various nuclidic composition, temperature and density. Assuming that $({}^{26}\text{Al}/{}^{27}\text{Al})_o = 1 \times 10^{-3}$, and substituting this value into eq. (5.17), Δt is evaluated as

$$\Delta t = (1.3 \pm 1.6) \times 10^6 \text{ years.}$$

This time interval is of the order of 10^5 to 10^6 years anyhow, and differs from what is called formation interval of 2×10^8 years estimated by ${}^{129}\text{I}$ - ${}^{129}\text{Xe}$ chronology. These two different time intervals suggest that at least two different nucleosynthetic events had formed those extinct isotopes. And they were injected into the primordial solar nebula.

5.3 ^{24}Mg ANOMALY

5.3.1 Yamato-75028

Data for matrix areas of Y-75028 chondrite are shown in Fig. 5.12. These data were taken in portions where the concentration ratio of Al/Mg was less than 0.13 in consideration that aluminum concentration is enough low to avoid the contribution of ^{26}Mg *. The plotted points concentrate around the origin and distribute near the normal mass fractionation line. From the results, it is judged there is no anomaly of magnesium.

5.3.2 Allende

Plot of $\Delta_{26/25}$ versus $\Delta_{24/25}$ for matrix portions of Allende is shown in Fig. 5.13. Plotted points correspond to the data for the portions where the concentration ratio of Al/Mg was less than 0.13 and Mg/Si ratio was in the range of 1.5 to 2.5. About 20 percent of the analyzed portions satisfied these ranges of concentration ratios.

Anomaly of $\Delta_{24/25}$ is distinctly shown in the figure. The maximum excess of ^{24}Mg is about 20 %.

5.3.3 Yamato-74191

The data for Y-74191 are shown in Fig. 5.14. These data were obtained for narrow matrix portions among chondrules, because the Y-74191 chondrite is almost occupied with chondrules as shown in the picture of Fig. 2.5. Al/Mg concentration ratios were less than 0.13 for all portions plotted as data. Two kinds of data are included based on the difference of Mg/Si ratios. One of them is for the portions where Mg/Si ratio is less than 1.3 and the other,

Mg/Si ratio is higher than 1.5

Plotted points for the higher Mg/Si portions show the clear excess of ^{24}Mg . For lower Mg/Si portions, some points show the clear anomaly of ^{24}Mg , but, as a whole, the anomaly is not remarkable compared with that for the higher Mg/Si portions.

5.3.4 Excess ^{24}Mg

The excess of ^{24}Mg observed for Mg-rich and Al-poor portions of matrices in AL and Y-74191 can be explained in two ways. One of them is as the result of the addition of practically pure ^{24}Mg to the pre-existed solar nebula, the other is as the result of the depletions of ^{25}Mg and ^{26}Mg by nearly the same fractions. The latter case is considered to be less possible to happen, because any nuclear processes which decrease both ^{25}Mg and ^{26}Mg by the same fractions can be rarely expected. Therefore, only the former case will be discussed in the following.

5.4 Consistent explanation of excesses of ^{24}Mg and ^{26}Mg

It is necessary to explain both excesses of ^{26}Mg in a white inclusion of Allende and of ^{24}Mg in matrix areas of Allende and Y-74191 consistently. As was described in section 5.2, ^{26}Al , which resulted in the excess of ^{26}Mg , has been considered successfully to be formed in a nucleosynthesis on the occasion of a supernova explosion. And this supernova is estimated to have exploded a few million years prior to the crystallization of the primordial minerals based on the ratio of $(^{26}\text{Al}/^{27}\text{Al})_c$ and the half life of ^{26}Al (7.2×10^5 y).

The formation interval from ^{129}I - ^{129}Xe was 2×10^8 years, and for the same meteorite, ^{26}Al - ^{26}Mg formation interval was $1-3 \times 10^6$ years. These two kinds of formation intervals are quite different with each other. If ^{129}I was formed in the late nucleosynthetic event, these two formation intervals should be the same values. Therefore, ^{129}I was not expected to be synthesized at the same time as the formation of ^{26}Al . The restriction leads to a prospect that neutron flux would be enough low in the case of the late nucleosynthetic event not to synthesize such r-process nuclei as ^{129}I . On the contrary, the explosion event of the previous supernova had synthesized ^{129}I through r-process under a condition of high neutron flux.

From the existence of the excess ^{24}Mg discovered for the matrices of Allende and Y-74191, it is thought that almost pure ^{24}Mg has been synthesized and injected into the pre-existed solar nebula.

This almost pure ^{24}Mg would be estimated to be synthesized through an explosive carbon burning process at the late supernova explosion according to the theoretical consideration by Arnett.³⁷⁾ Possible conditions of the supernova is as follows:

Composition: 50 % ^{12}C , 50 % ^{16}O

Temperature: 1.8×10^9 K

Density: 10^7 g/cm³

Among these conditions, temperature is the critical one. If the temperature is a little higher than this, for example 2×10^9 K, ^{24}Mg , ^{25}Mg and ^{26}Mg would have been formed in the abundances almost the same as the natural isotopic ones. Therefore, the discovery of the excess ^{24}Mg seems to rest-

dict the condition of the temperature of the late supernova. Moreover, the formations of almost pure ^{24}Mg and little ^{25}Mg and ^{26}Mg lead to the estimate that the late supernova explosion was of the type of low neutron flux, because the high neutron flux supernova must form not only ^{24}Mg but also ^{25}Mg and ^{26}Mg .

This low neutron condition is consistent with the aspect that r-process nucleus, ^{129}I , would not be synthesized in the late supernova explosion.

5.5 ^{20}Ne , ^{23}Na , and ^{28}Si

If the late supernova was exploded, ^{20}Ne , ^{23}Na and ^{28}Si would be also synthesized together with ^{16}O , ^{26}Al and ^{24}Mg . Among the elements of Ne, Na and Si, isotopic abundance anomaly of Si has been examined for the sample of Allende inclusions by Clayton and Mayeda,⁵⁰⁾ but only normal mass fractionations have been observed.

Since Na has only one stable isotope, it is impossible to detect the isotopic anomaly.

In case of Ne, it is possible to detect the anomalous ^{20}Ne in principle. But because the wide range variety of the abundance of ^{20}Ne has been observed in many chondrites, it would be much difficult to detect the anomalous ^{20}Ne , which might be injected together with ^{24}Mg and ^{16}O , of the order of permil.⁵¹⁾

6. CONCLUSION

The results stated in the previous section give the following points.

(1) Excess ^{26}Mg due to the in situ decay of now extinct ^{26}Al (half life = 7.2×10^5 y) has been detected in the Al-rich white inclusion in the Allende chondrite. The excess correlates with the concentration ratio of Al/Mg, and from the correlation, the ratio of $(^{26}\text{Al}/^{27}\text{Al})_c$ at the primordial mineral crystallization could be determined. The value of $(^{26}\text{Al}/^{27}\text{Al})_c$ was found to be $(2.8 \pm 2.2) \times 10^{-4}$ for this inclusion.

(2) The initial ratio of $(^{26}\text{Al}/^{27}\text{Al})_0$ at the time of nucleosynthesis was estimated to be approximately 1×10^{-3} based on the theoretical considerations by Arnett³⁷⁾, Truran and Cameron⁴⁸⁾ and Arnett and Wefel.⁴⁹⁾ Using the ratios of $(^{26}\text{Al}/^{27}\text{Al})_c$ and $(^{26}\text{Al}/^{27}\text{Al})_0$, a formation interval between the nucleosynthesis and the crystallization of the minerals could be estimated to be $(1.3 \pm 1.6) \times 10^6$ years. This value is distinctly different from the ^{129}I - ^{129}Xe formation interval which is 2×10^8 years.

(3) These results together with those reported by other workers on excess ^{26}Mg lead us to an explanation that at least two nucleosynthetic events had happened near the early solar nebula. The late event is considered to occur a few million years prior to the primordial mineral crystallization.

(4) The supernova explosion event would have formed ^{16}O , ^{20}Ne , ^{24}Mg , ^{26}Al and ^{28}Si through an explosive carbon burning process under a low neutron flux.

(5) Excess ^{24}Mg has been firstly found in Mg-rich and Al-poor portions in matrix areas of the Allende carbonaceous chondrite and the Yamato-74191 (L3) chondrite in the present work. The maximum excess of about 20 % has been observed in case of Allende.

(6) This excess of ^{24}Mg can be explained as the result of the addition of practically pure ^{24}Mg formed in the late supernova event to the pre-existed solar nebula.

(7) The excess of ^{24}Mg discovered in the present work, together with the excess of ^{16}O found by Clayton et al. and that of ^{26}Mg found by the author and the other workers, can be strong evidences which support the hypothesis that the primordial solar nebula was inhomogeneous and composed of at least two components.

APPENDIX

Software developed by the author himself for a precise isotopic ratio measurement is described. This software consists of three parts. One is for controlling a power supply of electromagnet, relays to switch high registers of an amplifier through interfaces. This is written by assembler. The second is for setting various initial conditions for measurement, and the third is for calculating isotopic ratios and for taking statistical procedures. These are written by BASIC. The lists of these programs are from the next page.

Disassembling list of the controlling program

6000 CDA467 M0	CALL IPR	6080 320481	STA SPFNT
6003 CDB767	CALL IIF	6083 E60F	ANI OFH
6006 CDC267	CALL ITM1	6085 FE07	CPI 07H
6009 CDB062	CALL OFREC	6087 CA9960	JZ R7
600C CD7562	CALL SLOW	608A FE08	CPI 08H
600F AF	XRA A	608C CAA160	JZ R8
6010 320281	STA SCCNT	608F FE09	CPI 09H
6013 210058	LXI H,PHSAV	6091 CAA960	JZ R9
6016 221081	SHLD PHPNT	6094 FE0A	CPI 0AH
6019 21005C	LXI H,TMSAV	6096 CAB160	JZ R10
601C 221281	SHLD TMPNT	6099 CD6663 R7	CALL ON7
601F CDF167	CALL ITM2	609C 1602	MVI D,02H
6022 CD7A62	CALL ONREC	609E C3B660	JMP T3
6025 1605	MVI D,05H	60A1 CD6C63 R8	CALL ON8
6027 3E00 TO	MVI A,00H	60A4 1602	MVI D,02H
6029 CD7B63	CALL TIM	60A6 C3B660	JMP T3
602C 15	DCR D	60A9 CD7163 R9	CALL ON9
602D C22760	JNZ TO	60AC 1603	MVI D,03H
6030 CDD267 M1	CALL IREG	60AE C3B660	JMP T3
6033 AF	XRA A	60B1 CD7663 R10	CALL ON10
6034 320381	STA PKCNT	60B4 1604 M2	MVI D,04H
6037 3A0281	LDA SCCNT	60B6 AF T3	XRA A
603A 3C	INR A	60B7 CD7B63	CALL TIM
603B 320281	STA SCCNT	60BA 15	DCR D
603E CD5162	CALL SCAON	60BB C2B660	JNZ T3
6041 3A0381 M11	LDA PKCNT	60BE CD3762	CALL ADHL
6044 3C	INR A	60C1 CDCC62	CALL BTB20
6045 320381	STA PKCNT	60C4 110800	LXI D,0008H
6048 2A1481	LHLD MXREG	60C7 19	DAD D
604B EB	XCHG	60C8 220AB1	SHLD BARGU
604C 2A1681	LHLD MNREG	60CB CDD267	CALL IREG
604F CDDE62	CALL INV	60CE AF	XRA A
6052 19	DAD D	60CF 320581	STA UDPNT
6053 EB	XCHG	60D2 3A0481	LDA SPFNT
6054 2A1081	LHLD PHPNT	60D5 E620	ANI 20H
6057 72	MOV M,D	60D7 CC7062	CZ FAST
6058 23	INX H	60DA C47562	CNZ SLOW
6059 73	MOV M,E	60DD 3E07 M4	MVI A,07H
605A 23	INX H	60DF 320781	STA COUNT
605B 221081	SHLD PHPNT	60E2 CD3762 M5	CALL ADHL
605E 2A1881	LHLD TMREG	60E5 CDCC62	CALL BTB20
6061 EB	XCHG	60E8 CD1C63	CALL MNMX
6062 2A1281	LHLD TMPNT	60EB EB	XCHG
6065 72	MOV M,D	60EC 2A0AB1	LHLD BARGU
6066 23	INX H	60EF CDDE62	CALL INV
6067 73	MOV M,E	60F2 19	DAD D
6068 23	INX H	60F3 7C	MOV A,H
6069 221281	SHLD TMPNT	60F4 E680	ANI 80H
606C 3A0381	LDA PKCNT	60F6 C2DD60	JNZ M4
606F 6F	MOV L,A	60F9 3A0781	LDA COUNT
6070 3A0081	LDA PKSET	60FC 3D	DCR A
6073 BD	CMP L	60FD 320781	STA COUNT
6074 DAE561	JC EO	6100 C2E260	JNZ M5
6077 2680	MVI H,80H	6103 3E20 M60	MVI A,20H
6079 23	INX H	6105 320781	STA COUNT
607A 7E	MOV A,M	6108 CD3762 M6	CALL ADHL
607B 320C81	STA SPFND	610B CDCC62	CALL BTB20
607E 2B	DCX H	610E CD1C63	CALL MNMX
607F 7E	MOV A,M	6111 EB	XCHG

continued

6112 C5	PUSH B	618C E680	ANI 80H
6113 0611	MVI B, 11H	618E CA0361	JZ M60
6115 0E00	MVI C, 00H	6191 3A0C81	LDA SPPND
6117 CD8562	CALL BCTBI	6194 E620	ANI 20H
611A C1	POP B	6196 CC7062	CZ FAST
611B CDDE62	CALL INV	6199 C47562	CNZ SLOW
611E 19	DAD D	619C C30861	JMP M6
611F 7C	MOV A, H	619F 3A0581 M8	LDA UDPNT
6120 E680	ANI 80H	61A2 FE00	CPI 00H
6122 CA0862	JZ 00	61A4 CACF61	JZ M10
6125 D5	PUSH D	61A7 3A0C81	LDA SPPND
6126 2A0A81	LHLD BARGU	61AA E620	ANI 20H
6129 111200	LXI D, 0012H	61AC CC7062	CZ FAST
612C 19	DAD D	61AF C47562	CNZ SLOW
612D CDDE62	CALL INV	61B2 2A0A81	LHLD BARGU
6130 D1	POP D	61B5 CDDE62	CALL INV
6131 19	DAD D	61B8 19	DAD D
6132 7C	MOV A, H	61B9 7C	MOV A, H
6133 E680	ANI 80H	61BA E680	ANI 80H
6135 C29F61	JNZ M8	61BC CA0361	JZ M60
6138 2A2A81	LHLD TRRG8	61BF CD7562	CALL SLOW
613B CDDE62	CALL INV	61C2 3A0781	LDA COUNT
613E 19	DAD D	61C5 3D	DCR A
613F 7C	MOV A, H	61C6 320781	STA COUNT
6140 E680	ANI 80H	61C9 C20861	JNZ M6
6142 C26C61	JNZ M7	61CC C34160	JMP M11
6145 7C	MOV A, H	61CF 2A2A81 M10	LHLD TRRG8
6146 FE00	CPI 00H	61D2 CDDE62	CALL INV
6148 C20861	JNZ M6	61D5 19	DAD D
614B 7D	MOV A, L	61D6 7C	MOV A, H
614C E6E0	ANI E0H	61D7 E680	ANI 80H
614E C20861	JNZ M6	61D9 C26C61	JNZ M7
6151 3A0581 M13	LDA UDPNT	61DC C30861	JMP M6
6154 FE01	CPI 01H	61DF 00	NOP
6156 CA0861	JZ M6	61E0 00	NOP
6159 3A0481	LDA SPPNT	61E1 00	NOP
615C E610	ANI 10H	61E2 00	NOP
615E CC7062	CZ FAST	61E3 00	NOP
6161 C47562	CNZ SLOW	61E4 00	NOP
6164 3E01	MVI A, 01H	61E5 3A0181 E0	LDA SCSET
6166 320581	STA UDPNT	61E8 47	MOV B, A
6169 C30861	JMP M6	61E9 3A0281	LDA SCCNT
616C D5 M7	PUSH D	61EC B8	CMP B
616D 2A1481	LHLD MXREG	61ED CAFE61	JZ E1
6170 EB	XCHG	61F0 CD3162	CALL KEY
6171 2A1681	LHLD MNREG	61F3 CA0362	JZ E2
6174 CDDE62	CALL INV	61F6 3AFC7D	LDA 7DFCH
6177 19	DAD D	61F9 3E03	MVI A, 03H
6178 0604	MVI B, 04H	61FB C30A62	JMP E3
617A CD5D63 M9	CALL HALF	61FE 3E00 E1	MVI A, 00H
617D 05	DCR B	6200 C30A62	JMP E3
617E C27A61	JNZ M9	6203 3E01 E2	MVI A, 01H
6181 EB	XCHG	6205 C30A62	JMP E3
6182 2A0A81	LHLD BARGU	6208 3E02 00	MVI A, 02H
6185 19	DAD D	620A 320681 E3	STA EPNT
6186 CDDE62	CALL INV	620D CD7163	CALL ON9
6189 D1	POP D	6210 CD7562	CALL SLOW
618A 19	DAD D	6213 3A0681	LDA EPNT
618B 7C	MOV A, H	6216 FE02	CPI 02H

continued

6218	CA2562	JZ	O1	6296	OF	RRC
621B	1606	MVI	D,06H	6297	E60F	BTB2 ANI OFH
621D	AF	T1	XRA A	6299	C2A562	JNZ BTB3
621E	CD7B63		CALL TIM	629C	B4	ORA H
6221	15		DCR D	629D	B5	ORA L
6222	C21D62		JNZ T1	629E	CAB662	JZ BTB5
6225	CD6162	O1	CALL SCAOF	62A1	AF	XRA A
6228	3A0681		LDA EPNT	62A2	C3AA62	JMP BTB4
622B	FE01		CPI 01H	62A5	FE0A	BTB3 CPI OAH
622D	C48062		CNZ OFREC	62A7	F2C662	JP BTB6
6230	C9		RET	62AA	54	BTB4 MOV D,H
6231	3AFE7D	KEY	LDA 7DFEH	62AB	5D	MOV E,L
6234	E620		ANI 20H	62AC	29	DAD H
6236	C9		RET	62AD	29	DAD H
6237	CD4362	ADHL	CALL ADDN	62AE	19	DAD D
623A	3E10		MVI A,10H	62AF	29	DAD H
623C	CD7B63		CALL TIM	62B0	85	ADD L
623F	2A047C		LHLD 7C04H	62B1	6F	MOV L,A
6242	C9		RET	62B2	7C	MOV A,H
6243	3E0B	ADON	MVI A,0BH	62B3	CE00	ACI 00H
6245	32077C		STA 7C07H	62B5	67	MOV H,A
6248	CD8963		CALL TIMB	62B6	F1	BTB5 POP PSW
624B	3E0A		MVI A,OAH	62B7	3F	CMC
624D	32077C		STA 7C07H	62B8	D28E62	JNC BTB1
6250	C9		RET	62BB	41	MOV B,C
6251	3E09	SCAON	MVI A,09H	62BC	3D	DCR A
6253	32037C		STA 7C03H	62BD	C28E62	JNZ BTB1
6256	3E50		MVI A,50H	62C0	F1	POP PSW
6258	CD7B63		CALL TIM	62C1	37	STC
625B	3E08		MVI A,0BH	62C2	3F	CMC
625D	32037C	SCA1	STA 7C03H	62C3	D1	POP D
6260	C9		RET	62C4	C1	POP B
6261	3E0B	SCAOF	MVI A,0BH	62C5	C9	RET
6263	32037C		STA 7C03H	62C6	F1	BTB6 POP PSW
6266	3E50		MVI A,50H	62C7	F1	POP PSW
6268	CD7B63		CALL TIM	62C8	37	STC
626B	3E0A		MVI A,OAH	62C9	D1	POP D
626D	C35D62		JMP SCA1	62CA	C1	POP B
6270	3E0D	FAST	MVI A,0DH	62CB	C9	RET
6272	C35D62		JMP SCA1	62CC	C5	BTB20 PUSH B
6275	3E0C	SLOW	MVI A,0CH	62CD	7C	MOV A,H
6277	C35D62		JMP SCA1	62CE	F5	PUSH PSW
627A	3E09	ONREC	MVI A,09H	62CF	E61F	ANI 1FH
627C	32077C	REC1	STA 7C07H	62D1	47	MOV B,A
627F	C9		RET	62D2	4D	MOV C,L
6280	3E08	OFREC	MVI A,0BH	62D3	CD8562	CALL BCTBI
6282	C37C62		JMP REC1	62D6	F1	POP PSW
6285	C5	BCTBI	PUSH B	62D7	E640	ANI 40H
6286	D5		PUSH D	62D9	CCDE62	CZ INV
6287	F5		PUSH PSW	62DC	C1	POP B
6288	210000		LXI H,0000H	62DD	C9	RET
628B	3E02		MVI A,02H	62DE	37	INV STC
628D	37		STC	62DF	3F	CMC
628E	F5	BTB1	PUSH PSW	62E0	7D	MOV A,L
628F	78		MOV A,B	62E1	2F	CMA
6290	D29762		JNC BTB2	62E2	C601	ADI 01H
6293	OF		RRC	62E4	6F	MOV L,A
6294	OF		RRC	62E5	7C	MOV A,H
6295	OF		RRC	62E6	2F	CMA

continued

62E7	CE00	ACI	00H			
62E9	67	MOV	H,A			
62EA	C9	RET				
62EB	C5	SUM	PUSH B			
62EC	D5		PUSH D			
62ED	210000	LXI	H,0000H			
62F0	011CB1	LXI	B,TRRG1			
62F3	3E0A	MVI	A,0AH			
62F5	F5	SUM1	PUSH PSW			
62F6	0A		LDAX B			
62F7	5F		MOV E,A			
62F8	03		INX B			
62F9	0A		LDAX B			
62FA	57		MOV D,A			
62FB	03		INX B			
62FC	19		DAD D			
62FD	F1		POP PSW			
62FE	3D		DCR A			
62FF	C2F562		JNZ SUM1			
6302	D1		POP D			
6303	C1		POP B			
6304	C9		RET			
6305	C5	ROTRG	PUSH B			
6306	D5		PUSH D			
6307	E5		PUSH H			
6308	0612		MVI B,12H			
630A	211CB1		LXI H,TRRG1			
630D	111EB1		LXI D,TRRG2			
6310	1A	ROTR1	LDAX D			
6311	77		MOV M,A			
6312	23		INX H			
6313	13		INX D			
6314	05		DCR B			
6315	C21063		JNZ ROTR1			
6318	E1		POP H			
6319	D1		POP D			
631A	C1		POP B			
631B	C9		RET			
631C	E5	MNMX	PUSH H			
631D	CD0563		CALL ROTRG			
6320	222EB1		SHLD TRRG1			
6323	CDEB62		CALL SUM			
6326	EB		XCHG			
6327	2A16B1		LHLD MNREG			
632A	CDDE62		CALL INV			
632D	19		DAD D			
632E	7C		MOV A,H			
632F	E680		ANI BOH			
6331	CA3A63		JZ MNMX1			
6334	EB		XCHG			
6335	2216B1		SHLD MNREG			
6338	E1		POP H			
6339	C9		RET			
633A	2A14B1	MNMX1	LHLD MXREG			
633D	CDDE62		CALL INV			
6340	19		DAD D			
6341	7C		MOV A,H			
6342	E680		ANI BOH			
6344	C25B63		JNZ MNMX2			
6347	EB		XCHG			
6348	2214B1		SHLD MXREG			
634B	3E80		MVI A,80H			
634D	320F7C		STA 7C0FH			
6350	3A0E7C		LDA 7C0EH			
6353	6F		MOV L,A			
6354	3A0E7C		LDA 7C0EH			
6357	67		MOV H,A			
6358	2218B1		SHLD TMREG			
635B	E1	MNMX2	POP H			
635C	C9		RET			
635D	37	HALF	STC			
635E	3F		CMC			
635F	7C		MOV A,H			
6360	1F		RAR			
6361	67		MOV H,A			
6362	7D		MOV A,L			
6363	1F		RAR			
6364	6F		MOV L,A			
6365	C9		RET			
6366	3E00	DN7	MVI A,00H			
6368	32007C	DN70	STA 7C00H			
636B	C9		RET			
636C	3E03	DN8	MVI A,03H			
636E	C36863		JMP DN70			
6371	3E05	DN9	MVI A,05H			
6373	C36863		JMP DN70			
6376	3E09	DN10	MVI A,09H			
6378	C36863		JMP DN70			
637B	C5	TIM	PUSH B			
637C	47		MOV B,A			
637D	0E00		MVI C,00H			
637F	0D	TIMO	DCR C			
6380	C27F63		JNZ TIMO			
6383	05		DCR B			
6384	C27F63		JNZ TIMO			
6387	C1		POP B			
6388	C9		RET			
6389	3E80	TIMB	MVI A,80H			
638B	3D	TIMBO	DCR A			
638C	C28B63		JNZ TIMBO			
638F	C9		RET			

continued

66FC	2A1081	K0	LHLD	PHPNT					
66FF	223081	K00	SHLD	PHTMP					
6702	C9		RET						
6703	2A1281	L0	LHLD	TMPNT					
6706	C3FF66		JMP	K00					
6709	2A3081	K1	LHLD	PHTMP					
670C	2B		DCX	H					
670D	7E		MOV	A,M					
670E	323381		STA	PHTM2					
6711	2B		DCX	H					
6712	7E		MOV	A,M					
6713	323281		STA	PHTM1					
6716	223081		SHLD	PHTMP					
6719	C9		RET						
671A	F5	NOLF	PUSH	PSW					
671B	3A7984		LDA	PRPNT					
671E	FE0D		CPI	ODH					
6720	CA2B67		JZ	NOLF1					
6723	FE0A		CPI	0AH					
6725	CA2B67		JZ	NOLF1					
6728	CD2F67		CALL	LPRI					
672B	F1	NOLF1	POP	PSW					
672C	C9		RET						
672D	3E0A	LFPR	MVI	A,0AH					
672F	320881	LPRI	STA	PRPN1					
6732	3A127C	PRINT	LDA	7C12H					
6735	E604		ANI	04H					
6737	C23267		JNZ	PRINT					
673A	3A0881		LDA	PRPN1					
673D	32107C		STA	7C10H					
6740	3E0C		MVI	A,0CH					
6742	32137C		STA	7C13H					
6745	3C		INR	A					
6746	32137C		STA	7C13H					
6749	C9		RET						
674A	F5	HDCPY	PUSH	PSW					
674B	3A7984		LDA	PRPNT					
674E	FE0D		CPI	ODH					
6750	CA5667		JZ	HDCP1					
6753	CD2F67		CALL	LPRI					
6756	F1	HDCP1	POP	PSW					
6757	C9		RET						
6758	00		NOP						
6759	00		NOP						
675A	00		NOP						
675B	00		NOP						
675C	3E07	BEL	MVI	A,07H					
675E	C32F67		JMP	LPRI					
6761	3E08	BS	MVI	A,08H					
6763	C32F67		JMP	LPRI					
6766	3E0B	VT	MVI	A,0BH					
6768	C32F67		JMP	LPRI					
676B	3E0C	FF	MVI	A,0CH					
676D	C32F67		JMP	LPRI					
6770	3E0D	CR	MVI	A,0DH					
6772	C32F67		JMP	LPRI					
6775	3E0E	SD	MVI	A,0EH					
6777	C32F67		JMP	LPRI					
677A	3E0F	SI	MVI	A,0FH					
677C	C32F67		JMP	LPRI					
677F	3E11	DC1	MVI	A,11H					
6781	C32F67		JMP	LPRI					
6784	3E12	DC2	MVI	A,12H					
6786	C32F67		JMP	LPRI					
6789	3E13	DC3	MVI	A,13H					
678B	C32F67		JMP	LPRI					
678E	3E14	DC4	MVI	A,14H					
6790	C32F67		JMP	LPRI					
6793	3E1B	ESC	MVI	A,1BH					
6795	C32F67		JMP	LPRI					
6798	00		NOP						
6799	00		NOP						
679A	00		NOP						
679B	00		NOP						
679C	CD9367	ESCE	CALL	ESC					
679F	3E45		MVI	A,45H					
67A1	C32F67		JMP	LPRI					
67A4	211A67	IPR	LXI	H,NOLF					
67A7	3EC3	I1	MVI	A,C3H					
67A9	32EC84		STA	84ECH					
67AC	22ED84		SHLD	84EDH					
67AF	21137C		LXI	H,7C13H					
67B2	3681		MVI	M,81H					
67B4	360D		MVI	M,0DH					
67B6	C9		RET						
67B7	3EB1	IIF	MVI	A,81H					
67B9	32037C		STA	7C03H					
67BC	3E93		MVI	A,93H					
67BE	32077C		STA	7C07H					
67C1	C9		RET						
67C2	210F7C	ITM1	LXI	H,7C0FH					
67C5	3634		MVI	M,34H					
67C7	3674		MVI	M,74H					
67C9	36B4		MVI	M,B4H					
67CB	C9		RET						
67CC	214A67	CHNG	LXI	H,HDCPY					
67CF	C3A767		JMP	I1					
67D2	160A	IREG	MVI	D,0AH					
67D4	CD3762	I2	CALL	ADHL					
67D7	CDCC62		CALL	BTB20					
67DA	CD0563		CALL	ROTRG					
67DD	222E81		SHLD	TRRGA					
67E0	15		DCR	D					
67E1	C2D467		JNZ	I2					
67E4	CDEB62		CALL	SUM					
67E7	221481		SHLD	MXREG					
67EA	21FF0F		LXI	H,0FFFH					
67ED	221681		SHLD	MNREG					
67F0	C9		RET						
67F1	210E7C	ITM2	LXI	H,7C0EH					
67F4	AF		XRA	A					
67F5	77		MOV	M,A					
67F6	77		MOV	M,A					
67F7	2B		DCX	H					
67F8	3632		MVI	M,32H					
67FA	77		MOV	M,A					
67FB	2B		DCX	H					
67FC	77		MOV	M,A					
67FD	3650		MVI	M,50H					
67FF	C9		RET						

List of BASIC controlling program

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5 CALL 17A4H: CALL 17B7H
10 DATA TI,HOG54,5,46,7.99,47,7.32,48,73.99,49,5.46,50,5.25
20 DATA SI,TAB(7),3,28,92.23,29,4.67,30,3.1
30 DATA MG,STW70,3,24,1,25,12663,26,139805,CU,TAB(7),2,63,69.2,65,30.8
40 DATA NI,BAR73,5,58,68,274,60,26,095,61,1,134,62,3,593,64,.904
50 DATA CR,ICAB0,4,50,4,35,52,83,79,53,9,5,54,2,36
60 DATA FE,JAM79,4,54,5,8,56,91,77,57,2,15,58,.28
70 DATA GE,TAB(7),5,70,20,5,72,27,4,73,7,8,74,36,5,76,7,8
80 DATA MO,TAB(7),5,94,9,3,95,15,9,96,16,7,97,9,6,98,24,1
90 DATA AG,TAB(7),2,107,51,83,109,48,17,0T,X,1,1,1
100 DIM A(5),B(5),C(5),D(5),E(5),F(25),H(5),I(5),J(5),K(5),L(5),M(4,5),N(50)
110 DIM P(25,5),Q(25,5),S(5),T(25,5),U(25,5),X(50,5),Y(50,5),Z(50,5)
120 LET A=1,B=0,D=A+A: CLEAR: PRINT "<AUTO SCAN>": PRINT
130 PRINT "INITIAL": GOSUB 1460: IF A$="Y" THEN POKE 80A0H,B
140 LET B$="C",V=B,E0=-120,P0=2.5,X3=5933,X4=34348,N4=PEEK(80A0H)
150 PRINT "POLARITY": PRINT "SCAN SPEED": PRINT "SENS.SWITCH": GOSUB 1460: IF
A$<>"Y" THEN 150
160 PRINT "": INPUT "DATE"N$
170 INPUT "NAME"D$: IF B$="N" THEN 830
180 INPUT "ELEMENT"F$: RESTORE
190 READ E$,G$,N: FOR I=A TO N: READ J(I),L(I): LET D(I)=J(I),E(I)=L(I): NEXT
I: IF E$="OT" THEN 230
200 IF E$<>F$ THEN 190
210 LET J1=N
220 CLEAR: PRINT G$: FOR I=A TO N: PRINT J(I);F$,"":L(I): NEXT I: GOSUB 146
0: IF A$="Y" THEN LET N1=N: GOTO 260
230 IF LEN(F$)>D THEN LET F$=LEFT(F$,D)
240 INPUT "PEAK NO."N: IF E$<>"OT" THEN GOSUB 1880: GOTO 220
250 FOR I=A TO N: INPUT "MASS"J(I),"ABUNDANCE"L(I): NEXT I: INPUT "REF."G$: GO
TO 220
260 INPUT "NORMALIZED TO MASS"T: FOR I=1 TO N: IF T=J(I) THEN LET E=I
270 NEXT I: LET E$=F$: FOR I=A TO N: LET K(I)=L(I)/L(E): NEXT I: POKE 8100H,N:
IF B$="T" THEN 830
280 PRINT "SELECT FROM (7,8,9,10)": FOR I=A TO N1
290 PRINT J(I);E$;: INPUT " SENSITIVITY="N: IF N>=7 THEN IF N<=10 THEN 310
300 GOTO 290
310 LET C=32768+I,A(I)=N: POKE C,N: NEXT I
320 GOSUB 1460: IF A$<>"Y" THEN 280

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continued

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330 LET H$="*PEAK",A1=A: GOSUB 1790: LET H$="*BASE",A1=D: GOSUB 1790
340 LET C=C+A: POKE C,B: IF B$="Z" THEN B30
350 INPUT "SCAN NO.(1<N<25)"N2: GOSUB 1460: IF A$<>"Y" THEN 350
360 LET N=N2+A: POKE 8101H,N: POKE 8102H,B: IF B$="X" THEN B30
370 INPUT "RUN NO."N5: GOSUB 1460: IF A$<>"Y" THEN 370
380 IF B$="Y" THEN B30
390 PRINT "INTERPO.AT": FOR I=A TO N1: PRINT J(I);E$: NEXT I: GOSUB 1490: FOR
I=A TO N1: IF N=J(I) THEN LET E1=I
400 NEXT I: IF B$="I" THEN B30
410 PRINT "ON-LINE": GOSUB 1460: LET N3=A: IF A$<>"Y" THEN LET N3=B
420 PRINT "GRAPH": GOSUB 1460: LET N7=A: IF A$<>"Y" THEN LET N7=D
430 PRINT "INITIAL SCAN POINT": GOSUB 1460: IF A$<>"Y" THEN 430
440 CLEAR : GOSUB 1850: PRINT "": PRINT "---START": GOSUB 1460: IF A$<>"Y" THEN
670
450 IF V=B THEN LET N6=A: GOTO 470
460 IF N6>A THEN LET N6=N6-A
470 IF V<>D THEN LET N4=N4+A: POKE 80A0H,N4
480 FOR J=B TO N1: LET X(N4,J)=B,Y(N4,J)=B: NEXT J: LET N(N4)=B: IF N3=B THEN
560
490 CALL 1784H: POKE X4,55H: CALL 1775H: GOSUB 1570: CALL X3: CALL 17A4H
500 CALL 17CCH: GOSUB 1470: CALL 17A4H: PRINT "(,G$,)": GOSUB 1480: IF N1>D+
D THEN CALL 177AH
510 PRINT E$," " : FOR I=A TO N1: PRINT #9,L(I): NEXT I: FOR I=A TO N1: IF I<
>E THEN PRINT #12,K(I)
520 NEXT I: CALL X3: CALL X3: PRINT " # " : FOR I=A TO N1: PRINT #9,J(I): NEX
T I: FOR I=A TO N1: IF I=E THEN 550
530 PRINT J(I);"/";J(E);: IF J(I)>99 THEN PRINT " ": GOTO 550
540 PRINT " "
550 NEXT I: CALL X3: CALL X3: POKE X4,56H
560 CLEAR : GOSUB 1570: PRINT "": GOSUB 1470: CURSOR 10,11: PRINT "< TOTAL > <
NOW >"
570 CURSOR A,13: PRINT "< RUN > ",#5,N5," ",N6
580 CURSOR A,15: PRINT "< SCAN > ",#5,N2: GOSUB 1500: IF V=B THEN CALL 1000
H: GOTO 600
590 CALL 1022H
600 LET V=PEEK(8106H): IF V=D THEN POKE 8102H,W: GOTO 650
610 IF N3=A THEN GOSUB 850
620 IF V=A+D THEN B30

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continued

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630 IF V=B THEN 940
640 GOSUB 1500: CALL 1030H: GOTO 600
650 CLEAR: PRINT "--OVER FLOW, DOWN EMT VOLT": PRINT
660 IF V<>B THEN PRINT "<C>CONT."
670 PRINT "<R>REPEAT": PRINT "<B>BUFFERING": PRINT "<S>STAT.": GOSUB 1850
680 PRINT "<L>LIST": PRINT "<U>SUMMARY": PRINT "<E>END": INPUT "WHICH" I$
690 FOR Q=A TO LEN(I$): LET B$=MID(I$,Q,A): IF B$="C" THEN 560
700 IF B$="R" THEN LET P0=2.5,E0=-120: POKE 8102H,B: GOTO 450
710 IF B$="B" THEN LET N(N4)=A: GOTO 830
720 IF B$="X" THEN 350
730 IF B$="Z" THEN 280
740 IF B$="N" THEN 170
750 IF B$="S" THEN 950
760 IF B$="T" THEN 180
770 IF B$="Y" THEN 370
780 IF B$="I" THEN 390
790 IF B$="U" THEN 1330
800 IF B$="L" THEN 1330
810 IF B$="E" THEN STOP
820 CLEAR: GOTO 660
830 IF V<>B THEN 820
840 NEXT Q: GOTO 820
850 CALL 16FCH: FOR I=N1 TO A STEP -A: GOSUB 1450: LET P(W,I)=(P1*256+P2)*INT(
10^(10-A(I))+.5): NEXT I
860 CALL 1703H: FOR I=N1 TO A STEP -A: GOSUB 1450: LET T(W,I)=P1*256+P2: NEXT
I: IF W=B THEN RETURN
870 LET E2=W: IF E1<>A THEN LET E2=W-A
880 FOR I=A TO N1
890 LET U(W,I)=P(W,I)+(T(E2,E1)-T(W,I))*(P(W-A,I)-P(W,I))/(T(W-A,I)-T(W,I)): N
EXT I
900 FOR I=A TO N1: LET Q(W,I)=U(W,I)/U(W,E): NEXT I
910 POKE X4,55H: PRINT #5,W: FOR I=A TO N1: PRINT #9,INT(U(W,I)+.5): NEXT I
920 FOR I=A TO N1: IF I<>E THEN PRINT #12,Q(W,I)
930 NEXT I: CALL X3: POKE X4,56H: RETURN
940 IF N3=B THEN 1310
950 CLEAR: PRINT "--STATISTICS, WAIT A MINUTE"
960 FOR I=A TO W: FOR J=A TO N1: LET P(I,J)=(Q(I,J)/K(J)-A)*1000: NEXT J: NEXT
I

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continued

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970 FOR I=A TO W: LET F(I)=B: NEXT I: LET L=B, Y=A
980 FOR J=A TO N1: LET S(J)=B, C(J)=B, K=B: FOR I=A TO W: IF F(I)=B THEN LET S(
J)=S(J)+Q(I, J), K=K+A
990 NEXT I: LET M(A, J)=S(J)/K, M(A+D, J)=(M(A, J)/K(J)-A)*1000: NEXT J
1000 FOR J=A TO N1: FOR I=A TO W: IF F(I)=B THEN LET C=Q(I, J)-M(A, J), C(J)=C(J)
+C*C
1010 NEXT I: LET C=C(J)/K, U=SQR(C)*D, M(D, J)=SQR(C/(K-A)), H(J)=M(A, J)+U, I(J)=M(A
, J)-U
1020 LET M(D+D, J)=M(D, J)/K(J)*1000: NEXT J: IF L=K THEN 1170
1030 POKE X4, 55H: IF N1>D+D THEN CALL 177AH
1040 PRINT "NO. OF DATA =", #4, K, "----*--": IF Y=A THEN 1070
1050 PRINT " (" : FOR I=A TO W: IF F(I)=Y-A THEN PRINT I; ", "
1060 NEXT I: PRINT ")"
1070 CALL X3: FOR J=A TO D+D: GOSUB 1540: ON J GOSUB 1510, 1520, 1530, 1520
1080 FOR I=A TO N1: IF I<E THEN PRINT #12, M(J, I)
1090 NEXT I: CALL X3: NEXT J: IF L=B THEN 1120
1100 FOR J=A TO N1: IF B(J)<M(D+D, J) THEN 1170
1110 NEXT J
1120 FOR I=A TO W: IF F(I)<>B THEN 1160
1130 FOR J=A TO N1: IF Q(I, J)<=H(J) THEN IF Q(I, J)>=I(J) THEN 1150
1140 LET F(I)=Y
1150 NEXT J
1160 NEXT I: FOR J=A TO N1: LET B(J)=M(D+D, J): NEXT J: LET L=K, Y=Y+A: GOTO 980
1170 GOSUB 1480: IF N6<N5 THEN IF V=B THEN CALL 127AH
1180 LET I=N4, M0=120, M1=-120: FOR J=A TO N1
1190 LET X(I, J)=M(A+D, J), Y(I, J)=M(D+D, J): IF M(A+D, J)<M0 THEN LET M0=M(A+D, J)
1200 IF M(A+D, J)>M1 THEN LET M1=M(A+D, J)
1210 LET Z(I, J)=J(J): NEXT J: LET X(I, B)=N1, Y(I, B)=E, P=2.5
1220 IF 10*P<M1-M0 THEN LET P=D*P: GOTO 1220
1230 IF P<P THEN LET P=P
1240 LET E3=P*(D-INT(M0/P)): IF E3>EO THEN LET EO=E3
1250 IF N7=D THEN 1300
1260 GOSUB 1530: CALL X3: CALL 177AH: GOSUB 1580: GOSUB 1590
1270 FOR I=A TO W: CALL 16E7H: FOR J=A TO N1: LET Z=P(I, J), C=64+J: IF F(I)<>B T
HEN LET C=63
1280 GOSUB 1600: NEXT J: CALL 16D0H: NEXT I: CALL X3: GOSUB 1590: CALL X3: CALL
16E7H: LET I=N4: GOSUB 1410
1290 GOSUB 1590: GOSUB 1580: POKE X4, 56H: CALL 176BH

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continued

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1300 IF V=A+D THEN B30
1310 IF N6>=N5 THEN B20
1320 LET N6=N6+A,V=B: POKE 8102H,B: GOTO 470
1330 INPUT "START #";N9,"END #";N8: CALL 177AH: LET E3=E0,P=P0
1340 POKE X4,55H: CALL 1775H: IF B#="U" THEN PRINT "SUMMARY (": GOTO 1360
1350 PRINT "LIST ("
1360 PRINT N9;"-";N8;" ) " ,N$: CALL X3: GOSUB 1620: IF B#="L" THEN 1400
1370 FOR I=N9 TO N8: CALL 16E7H: LET NO=N1,N1=X(I,B),E4=E,E=Y(I,B): GOSUB 1410:
LET N1=NO,E=E4
1380 IF N(I)=A THEN GOSUB 1590
1390 NEXT I: GOSUB 1590: GOSUB 1580: CALL 1784H: GOSUB 1530: CALL X3
1400 POKE X4,56H: CALL 176BH: GOTO B30
1410 FOR J=A TO N1: IF J=E THEN 1440
1420 LET C=45,Z=X(I,J)-Y(I,J)
1430 GOSUB 1600: LET Z=Z+P*.1: IF Z<=X(I,J)+Y(I,J) THEN 1430
1440 NEXT J: FOR J=A TO N1: LET Z=X(I,J),C=64+J: GOSUB 1600: NEXT J: CALL 16DOH
: RETURN
1450 CALL 1709H: LET P1=PEEK(8132H),P2=PEEK(8133H): RETURN
1460 INPUT "<Y/N>"A$: RETURN
1470 FOR I=A TO N1: PRINT J(I);E$;" = 10 E",#4,A(I): NEXT I: RETURN
1480 CALL 1784H: FOR I=A TO 13: PRINT "---*---": NEXT I: CALL X3: RETURN
1490 INPUT "WHICH ONE";N: RETURN
1500 LET W=PEEK(8102H): CURSOR 22,15: PRINT #4,W: CURSOR 30,15: RETURN
1510 PRINT " MEAN " : RETURN
1520 PRINT "ERROR " : RETURN
1530 PRINT "DELTA %." : RETURN
1540 LET F=9*N1-A-D
1550 FOR G=A TO F: PRINT " " : NEXT G: RETURN
1560 LET C=32768+I,A5=PEEK(C),B5=A5+N*16: POKE C,B5: RETURN
1570 PRINT N$," #",N4;"(",D$,")": RETURN
1580 FOR I=B TO 12*P STEP P: PRINT #10,I-E3: NEXT I: CALL X3: RETURN
1590 FOR J=A TO 13: PRINT "-----": NEXT J: CALL X3: RETURN
1600 LET N=32785+INT(10*(Z+E3)/P+.5): IF N>=32785 THEN IF N<=32917 THEN POKE
N,C
1610 RETURN
1620 GOSUB 1590: LET IO=N9: FOR H=N9 TO N8: LET S=X(H,B),E5=Y(H,B): IF IO<H TH
EN 1690
1630 PRINT " # " : FOR I=A TO D+D: GOSUB 1530: PRINT " +- " : GOSUB 1520: P
RINT " "

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continued

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1640 NEXT I: CALL X3: PRINT "      ": FOR I=A TO S: IF I=E5 THEN 1680
1650 PRINT Z(H,I);"/";Z(H,E5);: IF Z(H,I)>99 THEN LET F=15: GOTO 1670
1660 LET F=17
1670 GOSUB 1550
1680 NEXT I: CALL X3: CALL X3
1690 PRINT #8,H: FOR J=A TO S: IF J<>E5 THEN PRINT #12,X(H,J),Y(H,J)," "
1700 NEXT J: CALL X3: IF B#<>"L" THEN IF N(H)=A THEN GOSUB 1730
1710 NEXT H: LET H=H-A: IF N(H)=B THEN GOSUB 1730: CALL X3
1720 RETURN
1730 CALL X3: PRINT "G MEAN ": FOR J=A TO S: LET S(J)=B,C(J)=B: NEXT J
1740 FOR J=A TO S: IF J=E5 THEN 1780
1750 LET K=B: FOR R=IO TO H: LET S(J)=S(J)+X(R,J),K=K+A: NEXT R: LET M=S(J)/K
1760 FOR R=IO TO H: LET C=X(R,J)-M,C(J)=C(J)+C*K: NEXT R
1770 LET O=SQR(C(J)/K/(K-A)),X(B,J)=M,Y(B,J)=O: PRINT #12,M,O," "
1780 NEXT J: CALL X3: GOSUB 1590: LET IO=H+A: RETURN
1790 PRINT H#+" SPEED*": PRINT "<O>ALL FAST": PRINT "<1>ALL SLOW": PRINT "<2>EA
CH SET"
1800 GOSUB 1490: IF N>D THEN 1790
1810 IF N<>D THEN LET N=A1*N: FOR I=A TO N1: GOSUB 1560: NEXT I: RETURN
1820 CLEAR: PRINT "<O>FAST": PRINT "<1>SLOW": FOR I=A TO N1: IF A1=D THEN PRI
NT J(I)-A; "-";
1830 PRINT J(I): INPUT "SPEED=N: IF N>=D THEN 1820
1840 LET N=A1*N: GOSUB 1560: NEXT I: RETURN
1850 PRINT "<T>ELEMENT (";E#;")": PRINT "<N>NAME (";D#;")": PRINT "<X>SCAN #
(";N2;")"
1860 PRINT "<Y>RUN # (";N5;")": PRINT "<Z>SENS. (";: FOR I=A TO N1: PRINT A
(I);: NEXT I: PRINT ")"
1870 PRINT "<I>INTERPO.(";E1;")": RETURN
1880 FOR I=A TO N
1890 INPUT "MASS"JO: FOR J=A TO J1: IF JO=D(J) THEN LET J(I)=JO,L(I)=E(J): GOT
O 1910
1900 NEXT J: PRINT JO;"IS MISTAKE": GOTO 1890
1910 NEXT I: RETURN

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Table 2.1 Meteoritic samples used in the present work with their classifications and abbreviations

Sample	Classification	Abbreviation
Allende	C3	AL0, AL1, AL2
Yamato-74191	L3	Y-74191
Yamato-75028	H3	Y-75028

Table 2.2 Terrestrial samples used in the present work with their localities and abbreviations

Sample	Locality	Abbreviation
Forsterite in Dunite	Ehime Pref. Japan	FO
Olivine in Lherzolite	Salt Lake, Hawaii	SL46
Olivine in Lherzolite	Oki Island, Japan	OK
Olivine in Lherzolite	McMurdo, Antarctica	MM
Olivine in Spinel lherzolite	Salt Lake, Hawaii	SL45
Hornblende	Fukui Pref., Japan	HO
Vesuvianite	Chihuahua, Hawaii	VE
Cordierite	Kyoto Pref., Japan	CO
Anorthite	Hokkaido, Japan	AN
Feldspar in Granodiorite	Hyogo Pref., Japan	FE

Table 3.1 Typical operating conditions of the hollow cathode ion gun

	Hitachi IMA 2A	Home-made
Ionic species	Oxygen	Oxygen
Discharge voltage	400 V	1600 V
Discharge current	75 mA	30 mA

Table 3.2 Typical working conditions for magnesium isotopic analysis

	Hitachi IMA 2A	Home-made
Primary ion		
Energy	12-17 keV	8-9 keV
Beam diameter	70-200 μm	100-200 μm
Beam current	0.2-2 μA	0.2-0.8 μA
Secondary ion		
Accelerating voltage	3 kV	1 kV
Pressure		
Ultimate	3×10^{-5} Pa	6×10^{-5} Pa
during measurement	8×10^{-5} Pa	2×10^{-3} Pa

Table 3.3 Geometric parameters of mass spectrometers
and resolutions

	Hitachi IMA 2A	Home-made
Electrostatic sector		
Radius	15 cm	10 cm
Deflection angle	90°	63.6°
Magnetic sector		
Radius	12.5 cm	10 cm
Deflection angle	90°	90°
Slits		
Main slit	equal to beam dia.	$0.3 \times 4 \text{ mm}^2$
β slit	$0.5 \times 6 \text{ mm}^2$	$1.0 \times 6 \text{ mm}^2$
Collector slit	$0.5 \times 8 \text{ mm}^2$	$0.5 \times 4 \text{ mm}^2$
Resolution*	150	100

* Resolutions shown here are those for magnesium isotopic analysis.

Table 4.1 Interferences of possible molecular and doubly-charged ions to the subject ionic species

Subject ionic species	Possible interfering ionic species	Resolving* power	Maximum contribution to the subject peak	
			Home-made	Hitachi IMA
$^{24}\text{Mg}^+$	$^{12}\text{C}^{2+}$	1600	4×10^{-4}	3×10^{-5}
	$^{23}\text{NaH}^+$	1910	1×10^{-6}	2.5×10^{-6}
	$^{48}\text{Ca}^{2+}$	2730	4×10^{-4}	2×10^{-5}
	$^{48}\text{Ti}^{2+}$	2170	6×10^{-5}	7×10^{-5}
$^{25}\text{Mg}^+$	$^{12}\text{C}^{13}\text{C}^+$	1430	8×10^{-5}	5×10^{-6}
	$^{12}\text{C}_2\text{H}^+$	1140		1×10^{-5}
	$^{23}\text{NaH}_2^+$	1280		1.5×10^{-4}
	$^{50}\text{Ti}^{2+}$	1860	3×10^{-5}	4×10^{-5}
	$^{50}\text{Cr}^{2+}$	1950	6×10^{-5}	1×10^{-6}
	$^{24}\text{MgH}^+$	3560	1×10^{-3}	4×10^{-4}
$^{26}\text{Mg}^+$	$^{13}\text{C}_2^+$	1080	8×10^{-5}	7×10^{-6}
	$^{12}\text{C}^{14}\text{N}^+$	1270		
	$^{12}\text{C}^{13}\text{CH}^+$	910		
	$^{12}\text{C}_2\text{H}_2^+$	790	5×10^{-4}	5×10^{-4}
	$^{52}\text{Cr}^{2+}$	2110		
	$^{25}\text{MgH}^+$	2350	3×10^{-4}	5×10^{-5}

* Resolving power necessary to resolve an interfering ionic species from the subject one.

Table 4.2 Hydride ion formation ratio in cases without
and with a cold finger of liquid nitrogen
aside the sample holder

Liq. N ₂	$^{24}\text{MgH}^+ / ^{24}\text{Mg}^+ (=x)$	$^{24}\text{MgH}^+ / ^{25}\text{Mg}^+$
without	$\sim 4 \times 10^{-3}$	$\sim 3 \times 10^{-2}$
with	$< 5 \times 10^{-5}$	$< 4 \times 10^{-4}$

Table 4.3 Secondary ion formation ratios of $^{12}\text{C}_2^+ / ^{12}\text{C}^+$
and $(^{13}\text{C}_2^+ + ^{12}\text{C}^{14}\text{N}^+ + ^{12}\text{C}^{13}\text{CH}^+ + ^{12}\text{C}_2\text{H}_2^+) / ^{12}\text{C}^+$ for five
carbon-containing terrestrial samples

Sample	$^{12}\text{C}_2^+ / ^{12}\text{C}^+$	$Y^* / ^{12}\text{C}^+$
Graphite	0.054	0.0013
Calcite	0.003	0.0001
Silicon carbide	0.016	0.0004
Dolomite	~ 0	~ 0
Charcoal	0.040	0.0010

$$* Y = (^{13}\text{C}_2^+ + ^{12}\text{C}^{14}\text{N}^+ + ^{12}\text{C}^{13}\text{CH}^+ + ^{12}\text{C}_2\text{H}_2^+)$$

Table 4.4 Concentrations of Mg, Al and Si in four mineral samples
determined by atomic absorption analysis (wt.%) and
chemical formulae of the minerals

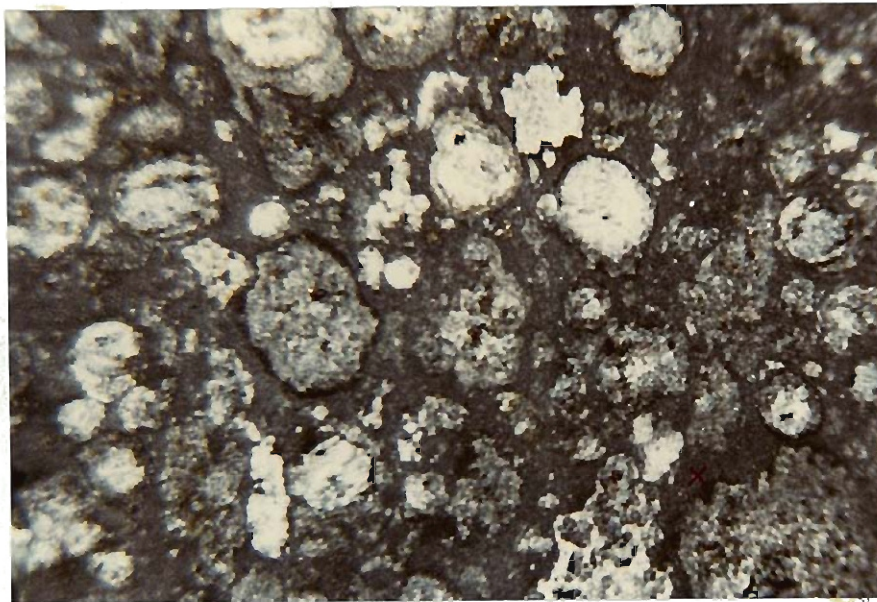
Sample	Mg	Al	Si	Chemical formula
Forsterite	31.1	0.2	16.9	Mg_2SiO_4
Hornblende	6.2	7.3	18.4	$(\text{Ca}, \text{Na})_{2-3}(\text{Mg}, \text{Fe}, \text{Al})_5[\text{OH} \cdot (\text{Si}, \text{Al})_{11}]_2$
Vesuvianite	2.7	8.0	15.4	$\text{Ca}_{10}(\text{Mg}, \text{Fe})_2\text{Al}_4[(\text{OH})_4 \cdot (\text{SiO}_4)_5 \cdot (\text{Si}_2\text{O}_7)_2]$
Cordierite	1.3	13.3	20.2	$\text{Mg}_2\text{Al}_3[\text{AlSi}_5\text{O}_{18}]$

Table 4.5 Isotopic ratios of magnesium obtained for the sample of terrestrial forsterite (FO) and used as the reference values

	Home-made	Hitachi IMA 2A
$^{25}\text{Mg}/^{24}\text{Mg}$	0.12495	0.12472
$^{26}\text{Mg}/^{24}\text{Mg}$	0.13666	0.13560
$^{24}\text{Mg}/^{25}\text{Mg}$	8.0032	8.0178
$^{26}\text{Mg}/^{25}\text{Mg}$	1.0937	1.0872

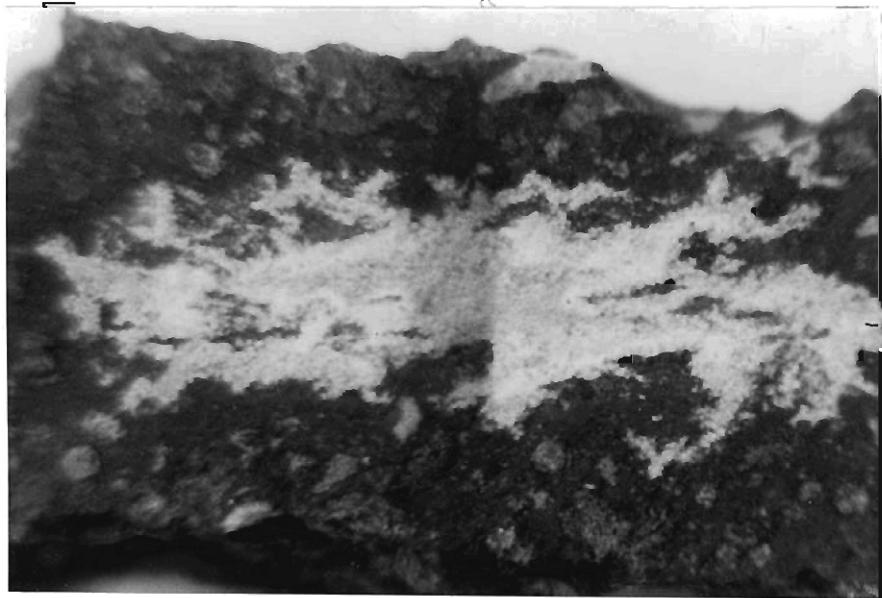
Table 5.1 $^{26}\text{Al}/^{27}\text{Al}$ ratio at the time of nucleosynthesis, $(^{26}\text{Al}/^{27}\text{Al})_0$, based on theoretical investigations

$(^{26}\text{Al}/^{27}\text{Al})_0$	Reference
1.2×10^{-3}	R.A. Fish et al. (1960) ⁵⁾
3.3×10^{-4}	W.A. Fowler et al. (1962) ²⁾
6.6×10^{-6}	H. Reeves and J. Audouze (1968) ⁶⁾
9.0×10^{-4}	W.D. Arnett (1969) ³⁷⁾
1.3×10^{-3}	W.D. Arnett (1969) ³⁷⁾
$0.4\text{-}2.0 \times 10^{-3}$	J.W. Truran and A.G.W. Cameron (1978) ⁴⁸⁾
$1\text{-}2 \times 10^{-3}$	W.D. Arnett and J.P. Wefel (1978) ⁴⁹⁾



1 mm

Fig. 2.1 Photomicrograph of a cut surface of the Allende carboaceous chondrite.



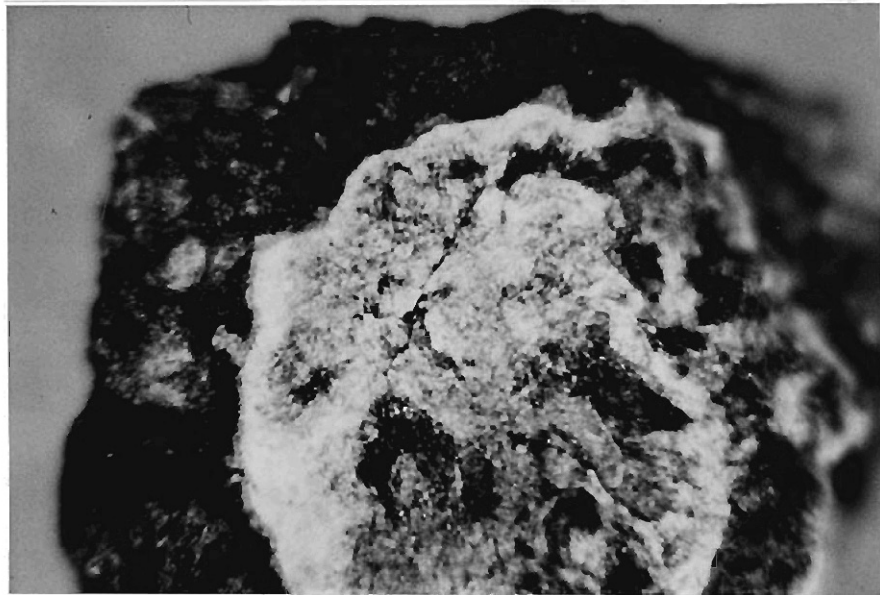
┌───┐
1 mm

Fig. 2.2 Photomicrograph of a cut surface of Allende including an amoeboid whitish inclusion.



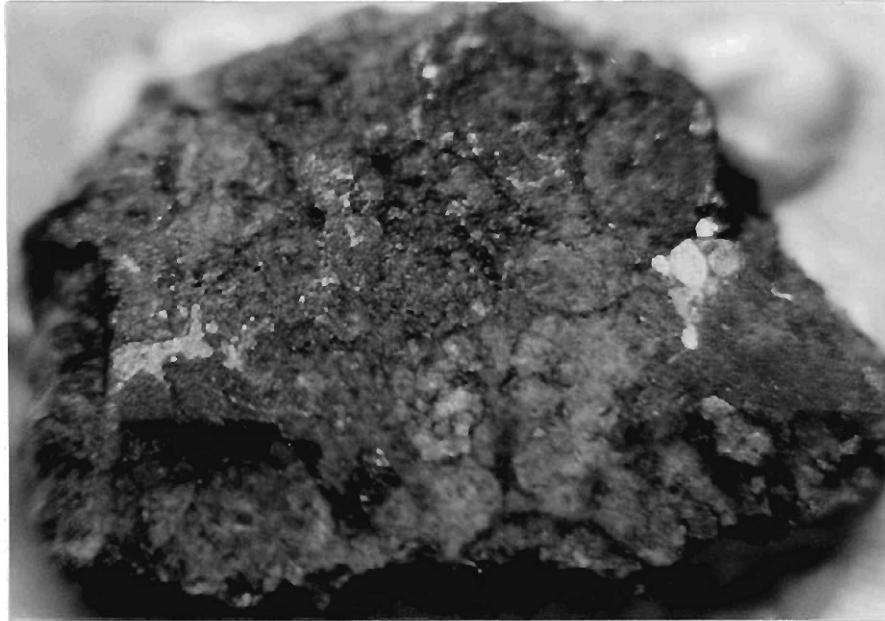
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1 mm

Fig. 2.3 Photomicrograph of a cut surface of Allende including a chondrule-like white inclusion surrounded by ring-shaped boundary layer.



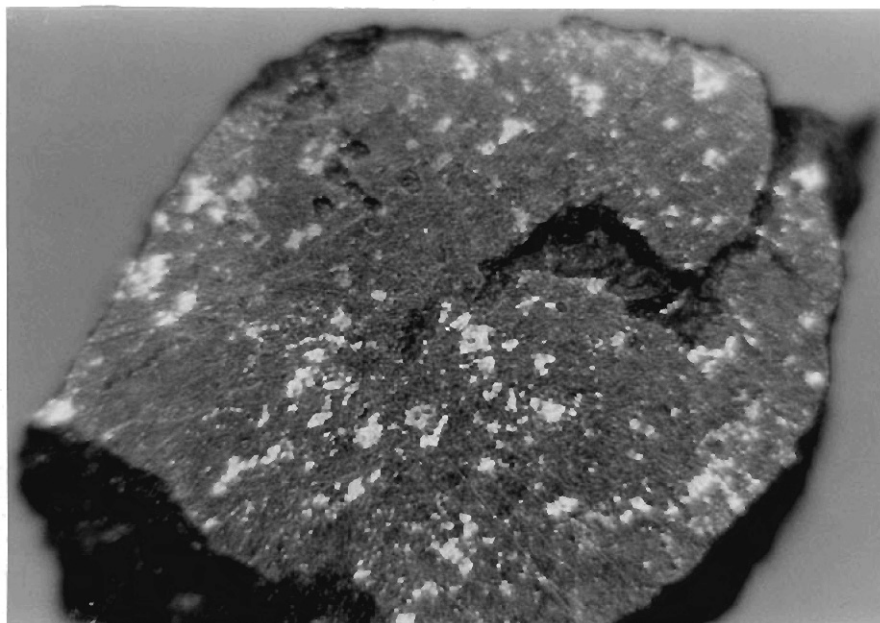
1 mm

Fig. 2.4 Photomicrograph of a cut surface of Allende including a large white inclusion.



1 mm

Fig. 2.5 Photomicrograph of a cut surface of Yamato-74191 (L3) chondrite.



┌───┐
1 mm

Fig. 2.6 Photomicrograph of a cut surface of Yamato-75028 brecciated chondrite. H3 part is taken in this picture.

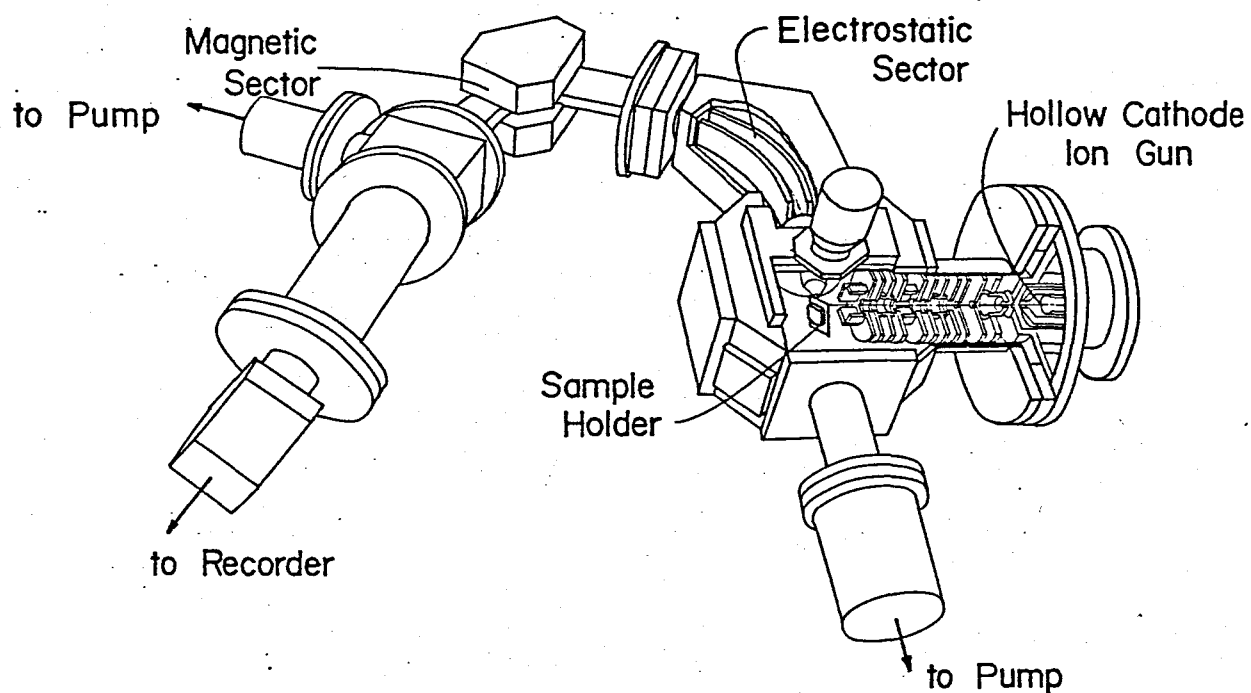


Fig. 3.1 Schematic diagram of a home-made ion microprobe mass analyzer.

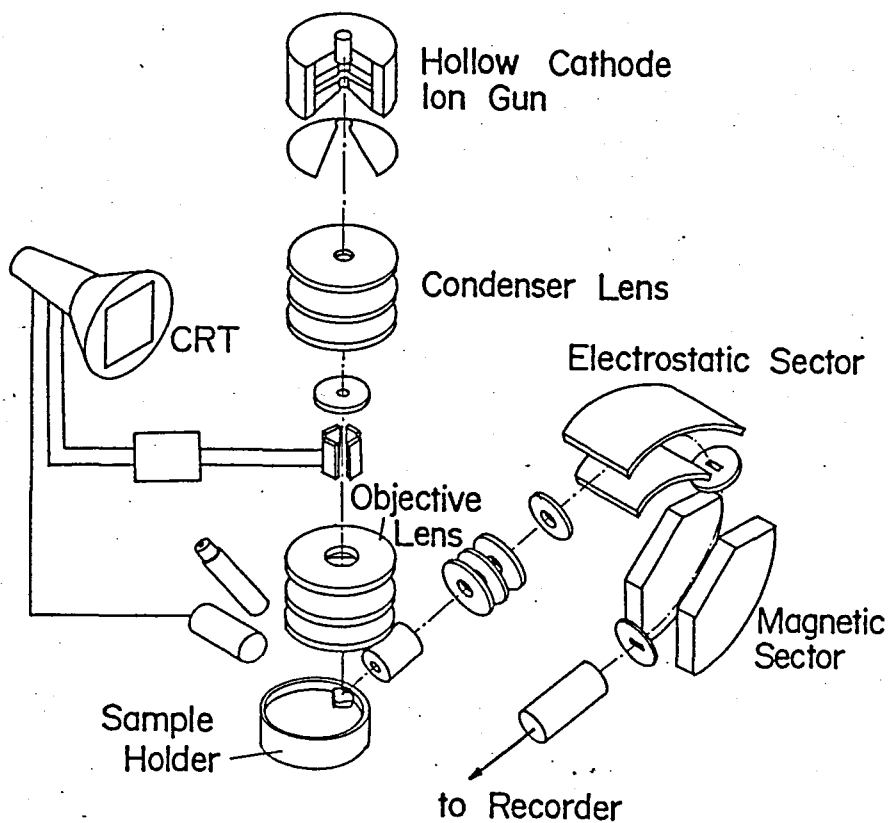


Fig. 3.2 Schematic diagram of a Hitachi IMA 2A ion microprobe mass analyzer.

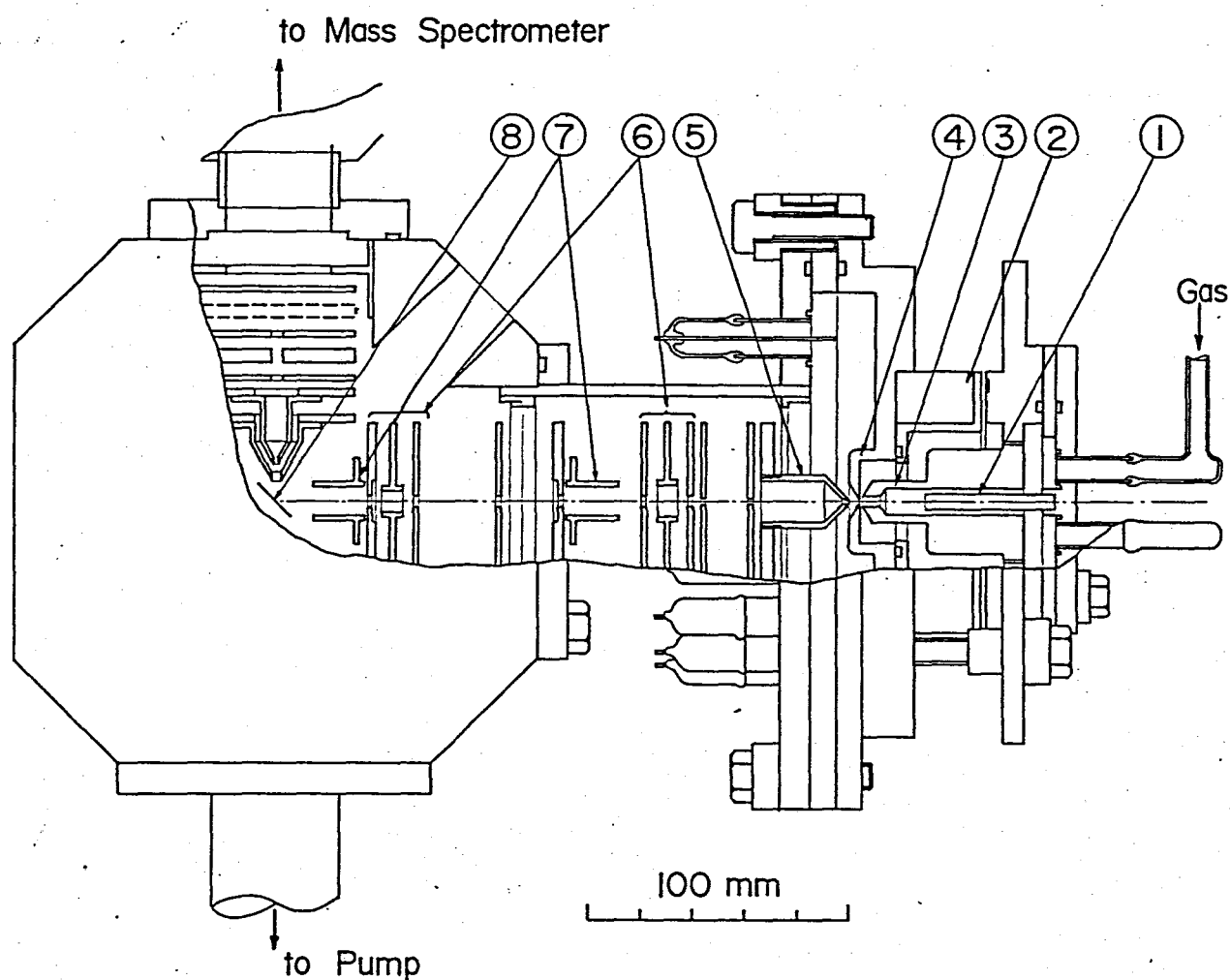


Fig. 3.3 Schematic diagram of a primary ion gun and accelerating and focusing system for the primary ion beam and for the secondary ions.

- ① hollow cathode
- ② ferrite permanent magnet
- ③ intermediate electrode
- ④ anode
- ⑤ drawing out and accelerating electrode
- ⑥ Einzel lens
- ⑦ deflector
- ⑧ sample holder

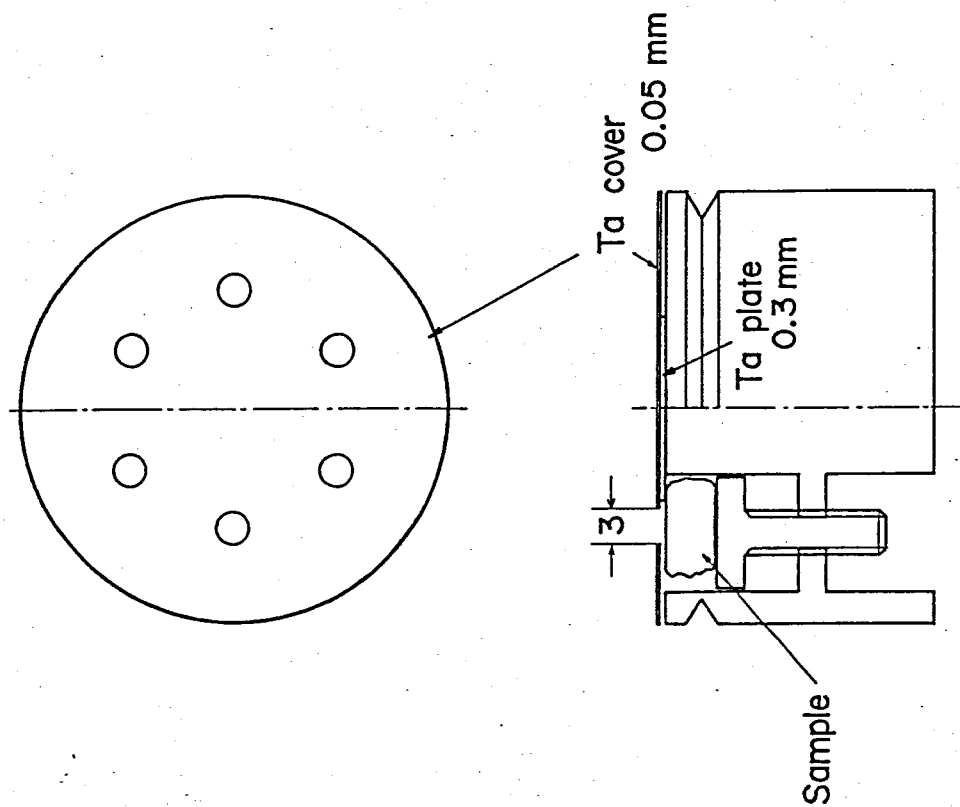


Fig. 3.5 Sample mounting system of the Hitachi IMA 2A apparatus.

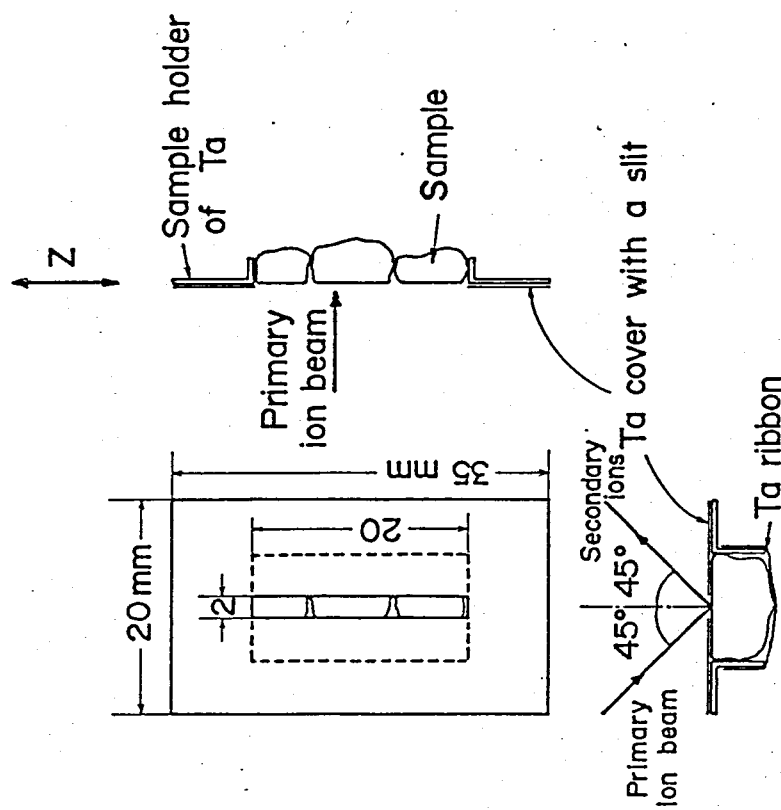


Fig. 3.4 Sample mounting system of the home-made apparatus. This is movable in the Z direction.

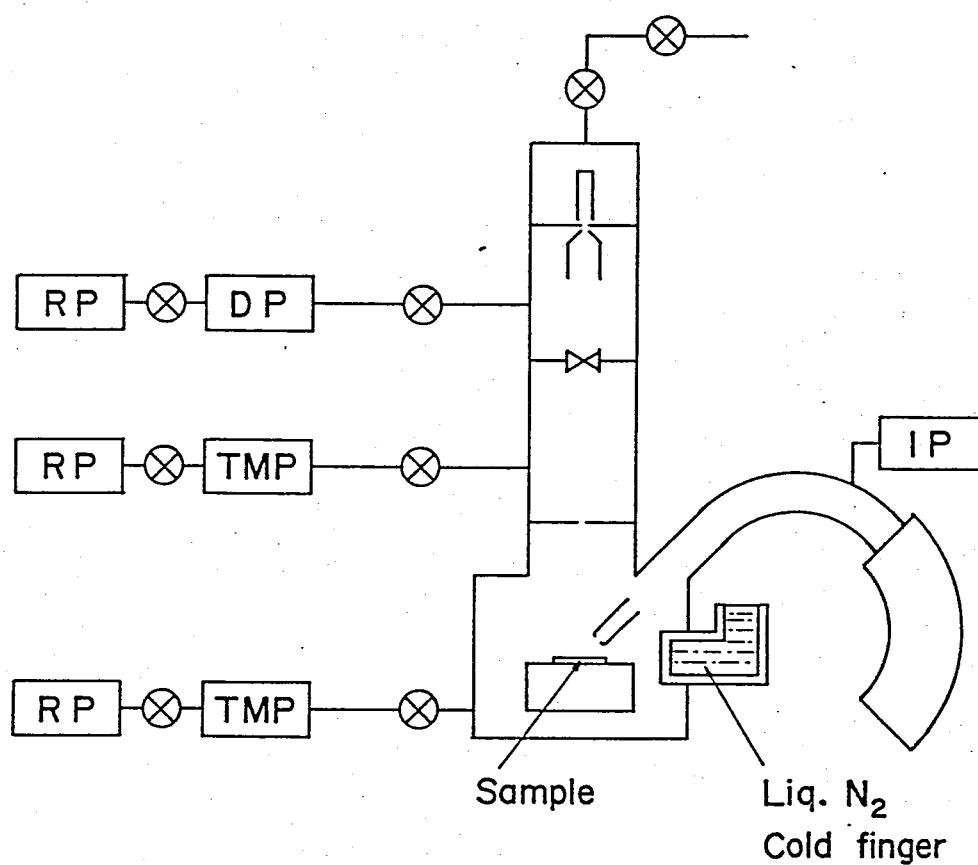


Fig. 3.6 Schematic diagram of the installation of a cold finger of liquid nitrogen, and pumping systems.

RP: Oil rotary pump, DP: Oil diffusion pump, TMP: Turbo-molecular pump, IP: Ion pump

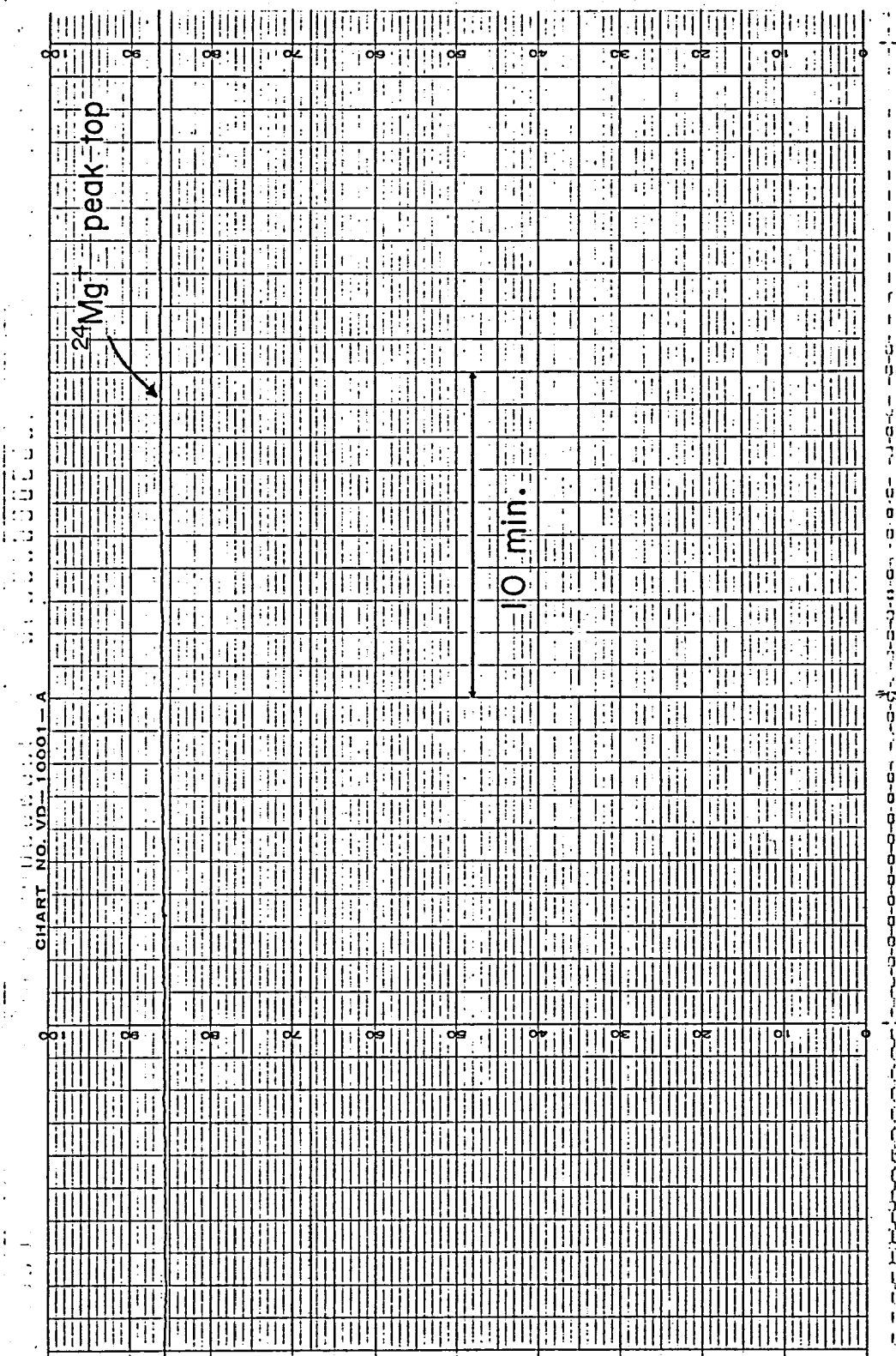


Fig. 3.7 Chart recording the peak top of $^{24}\text{Mg}^+$ for a terrestrial forsterite sample. This shows the stability of secondary ion current.

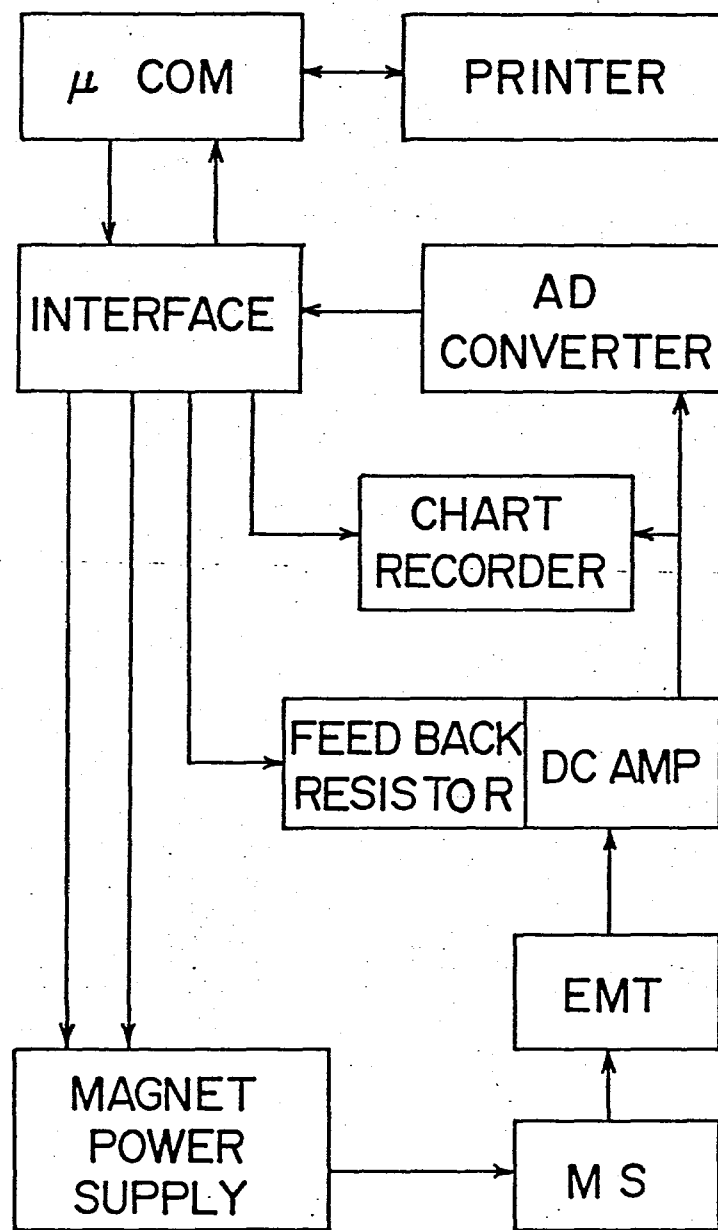


Fig. 3.8 Block diagram of a controlling system using a microcomputer.

μCOM: microcomputer with 8080AFC as CPU

EMT: secondary electron multiplier

MS: mass spectrometer

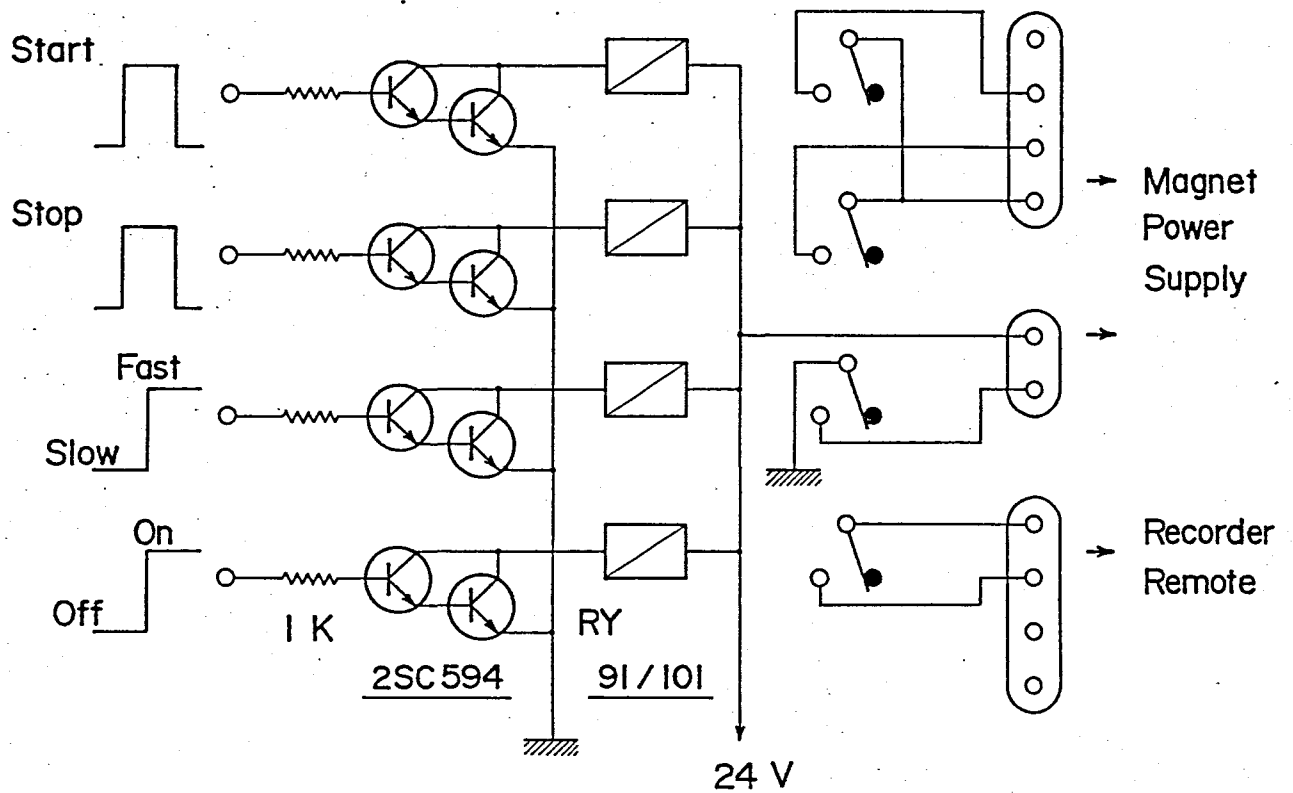


Fig. 3.9 Interfaces for a magnet power supply and a chart recorder.

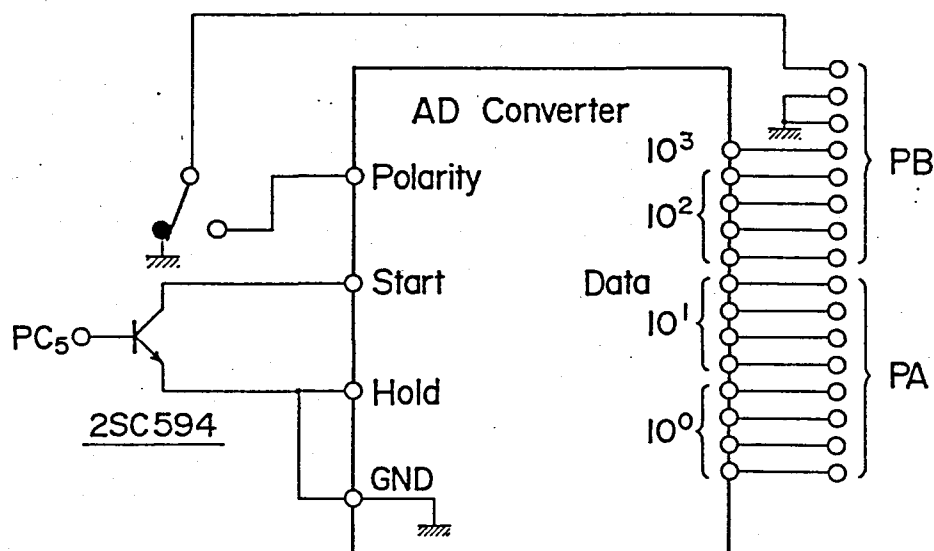


Fig. 3.10 Diagram of connections between an AD converter and a microcomputer.

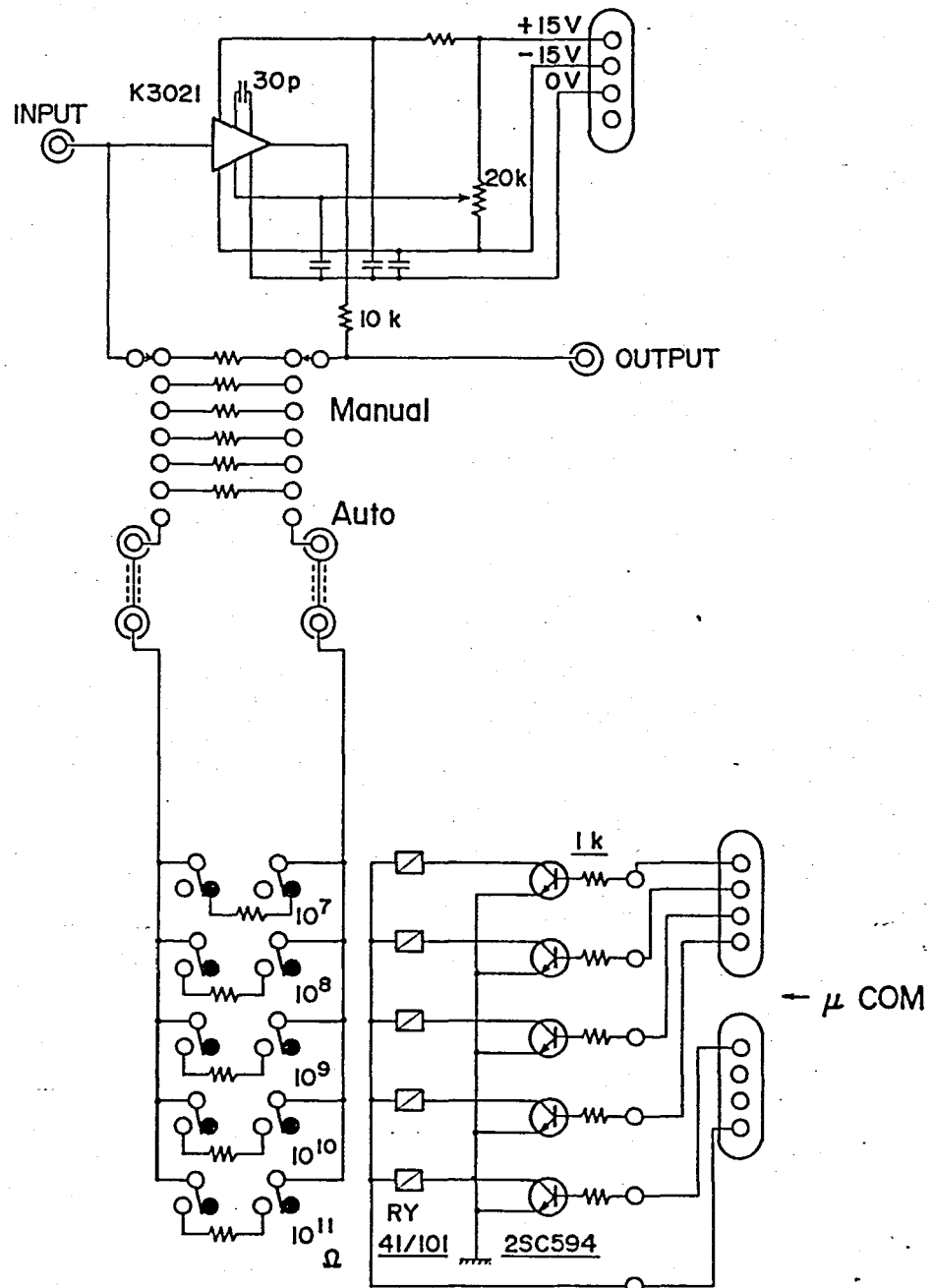


Fig. 3.11 Circuit for switching sensitivities of detecting system by switching a feed back resistor of an amplifier (K3021).

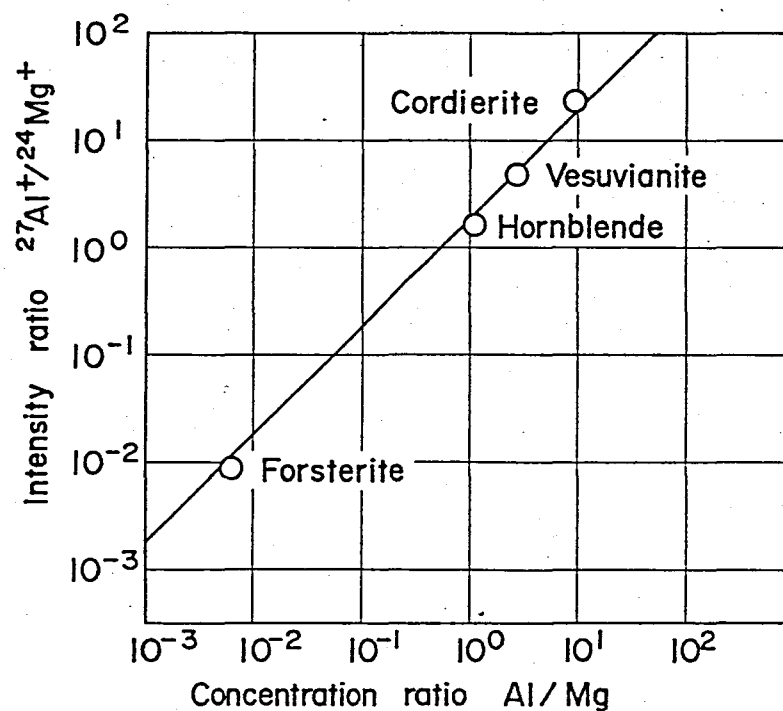


Fig. 4.1 Calibration curve for Al/Mg ratio. Elemental concentration was measured by atomic absorption analysis. The localities and the chemical formulae of the minerals are shown in Tables 2:2 and 4:4.

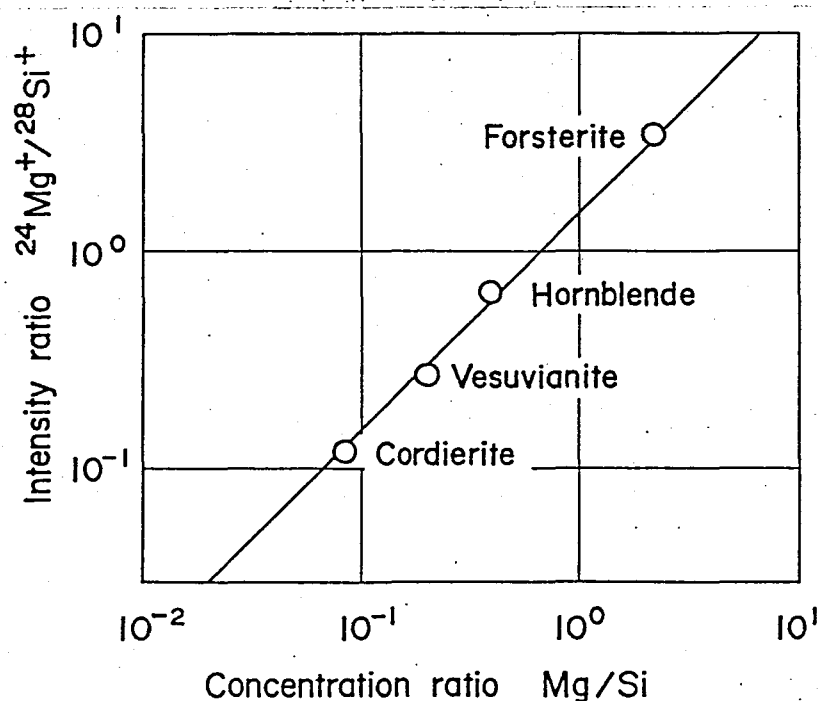


Fig. 4.2 Calibration curve for Mg/Si ratio. Elemental concentration was measured by atomic absorption analysis. The localities and the chemical formulae of the minerals are shown in Tables 2:2 and 4:4.

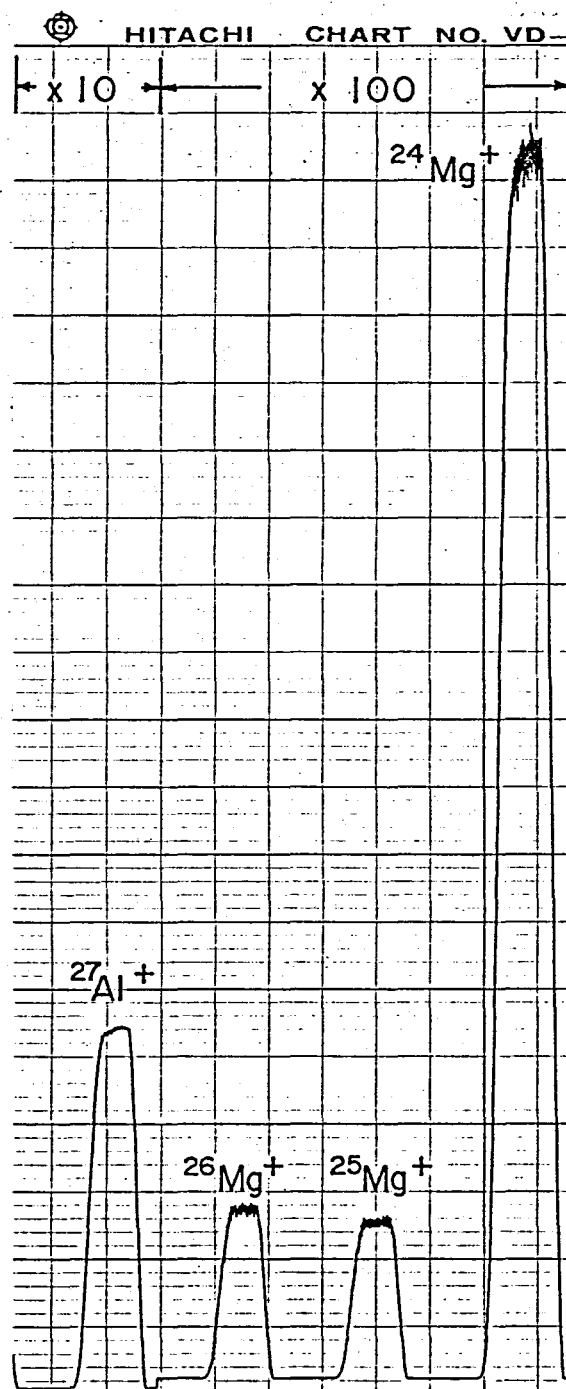


Fig. 4.3 Mass spectrum obtained for the white inclusion AL2 with the home-made apparatus. The sensitivities are shown in the upper part.

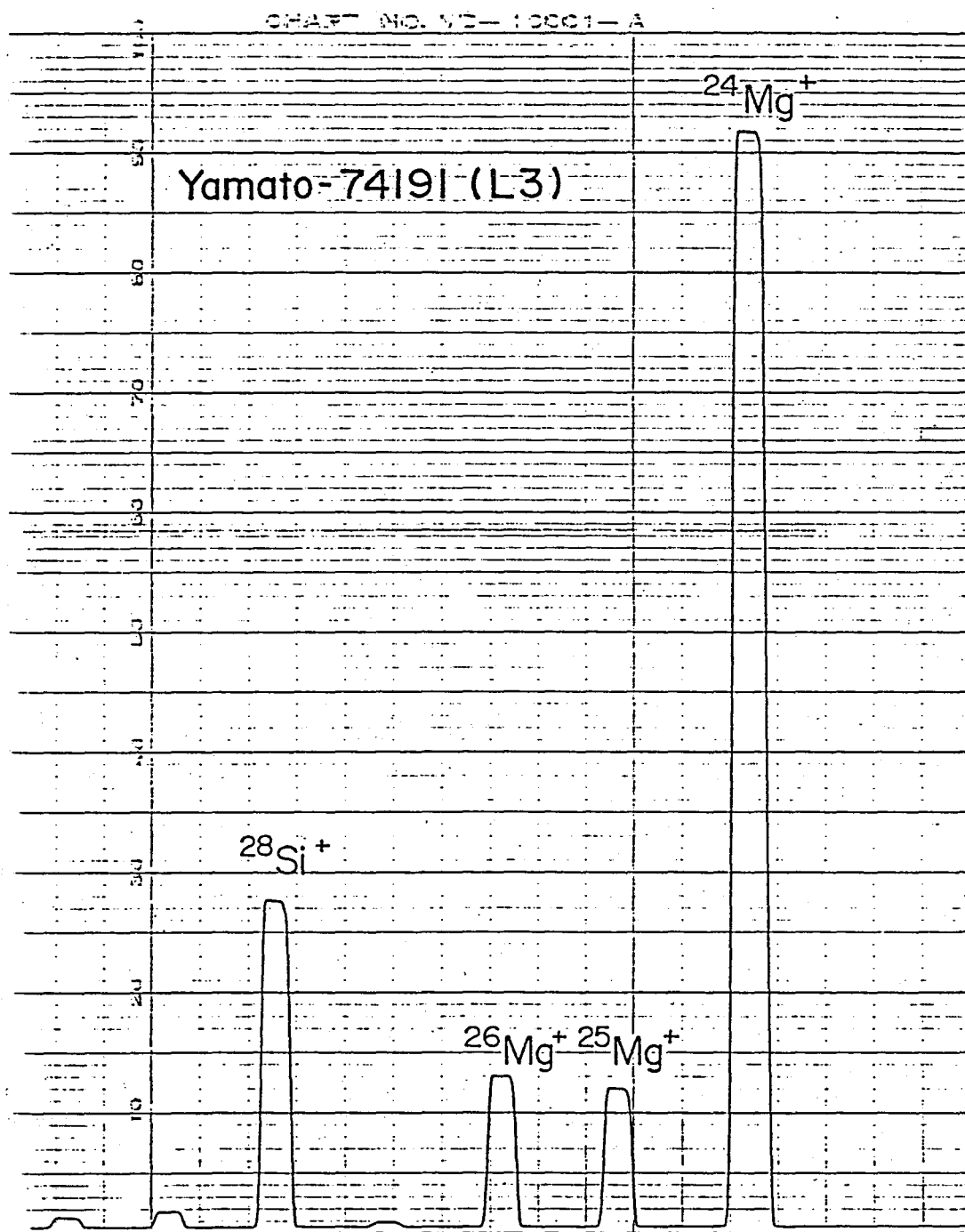


Fig. 4.4 Mass spectrum obtained for a portion of the matrix of Yamato-74191 chondrite with the Hitachi IMA 2A apparatus.

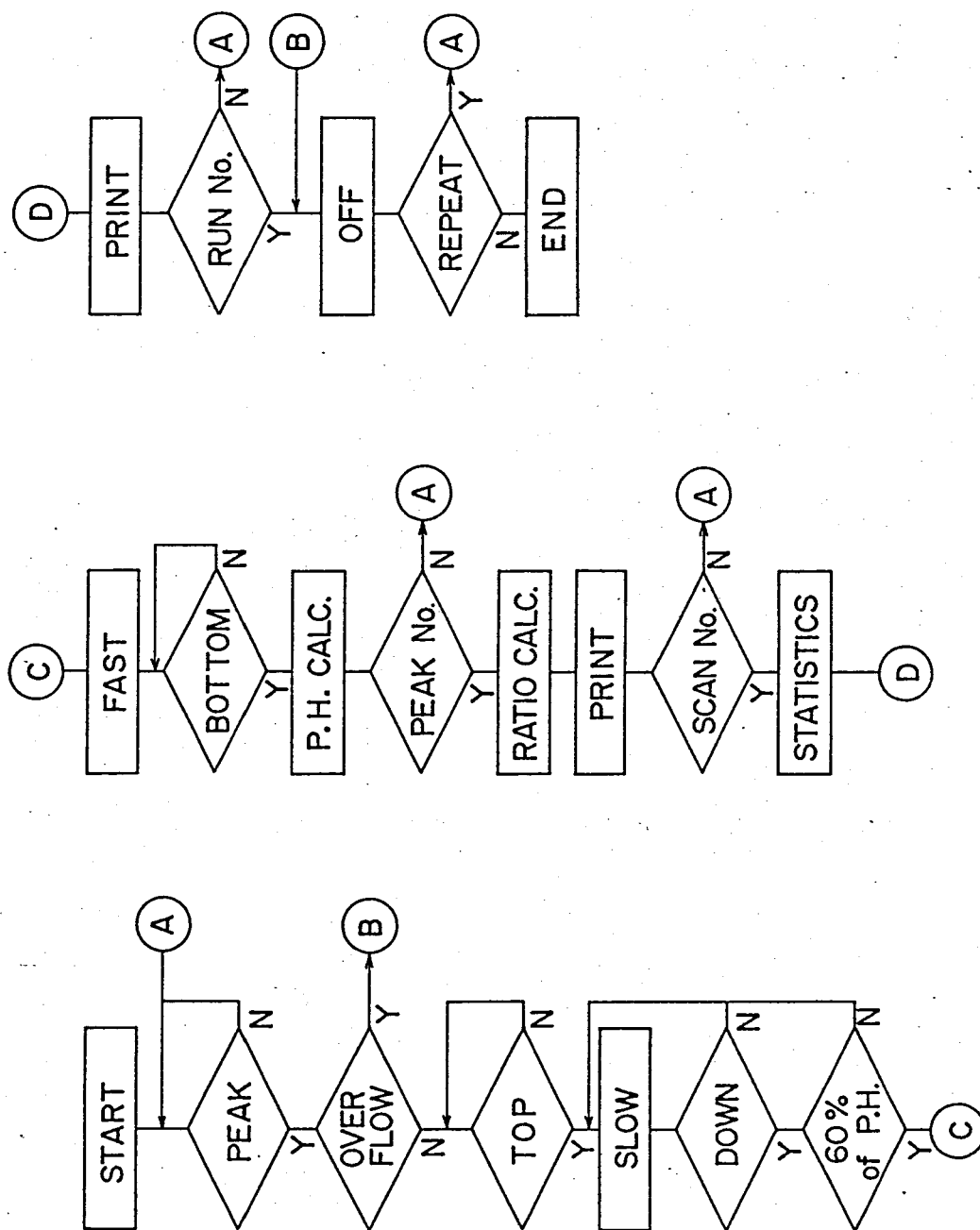


Fig. 4.5 Flow chart of obtaining isotopic ratios by using a microcomputer.

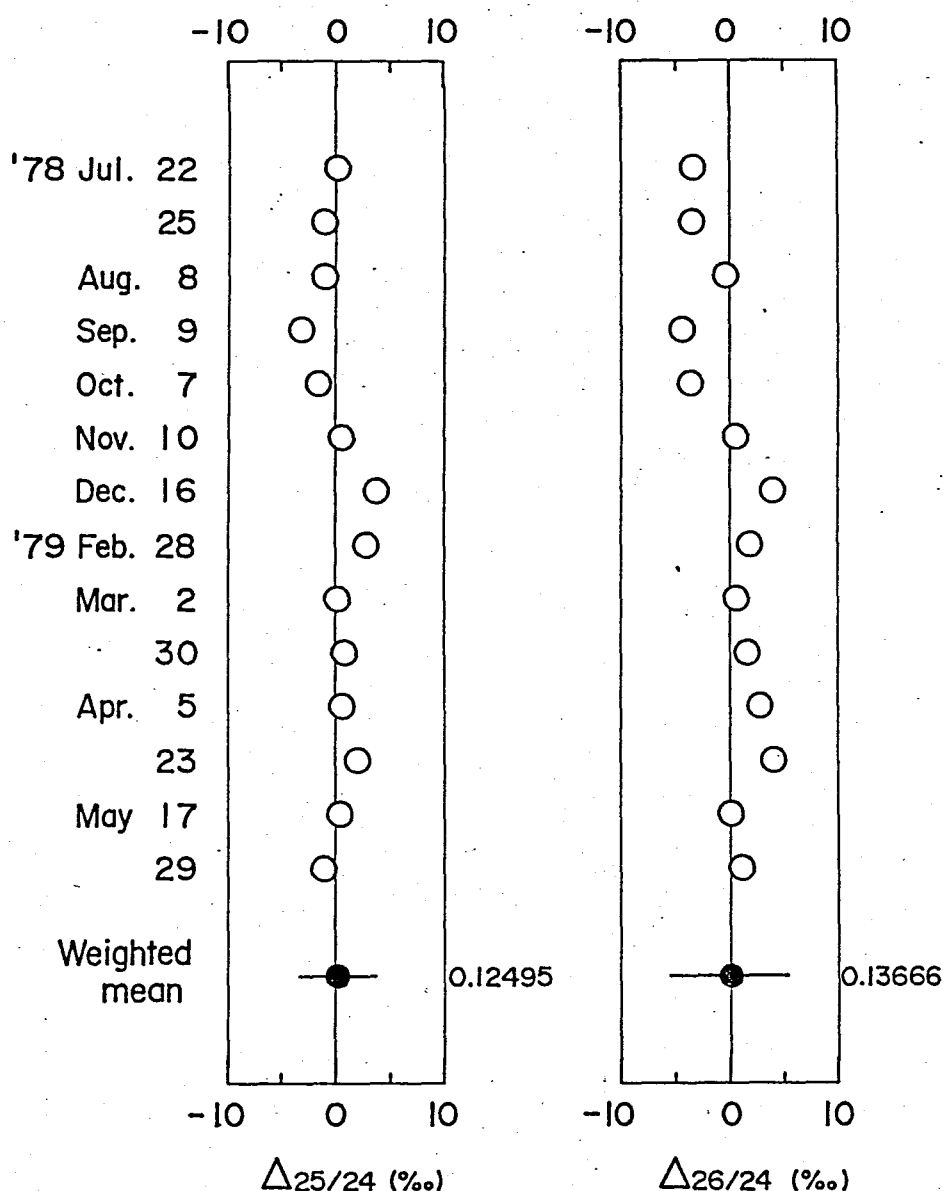


Fig. 4.6 Δ values obtained for the laboratory standard sample of terrestrial forsterite (F0). These data are plotted in the chronological order. The definition of Δ appears in the text. Values aside the closed circles are the weighted means of all these data, and these values were used as reference values in evaluating Δ values. Error bars for the closed circles represent twice the standard deviations.

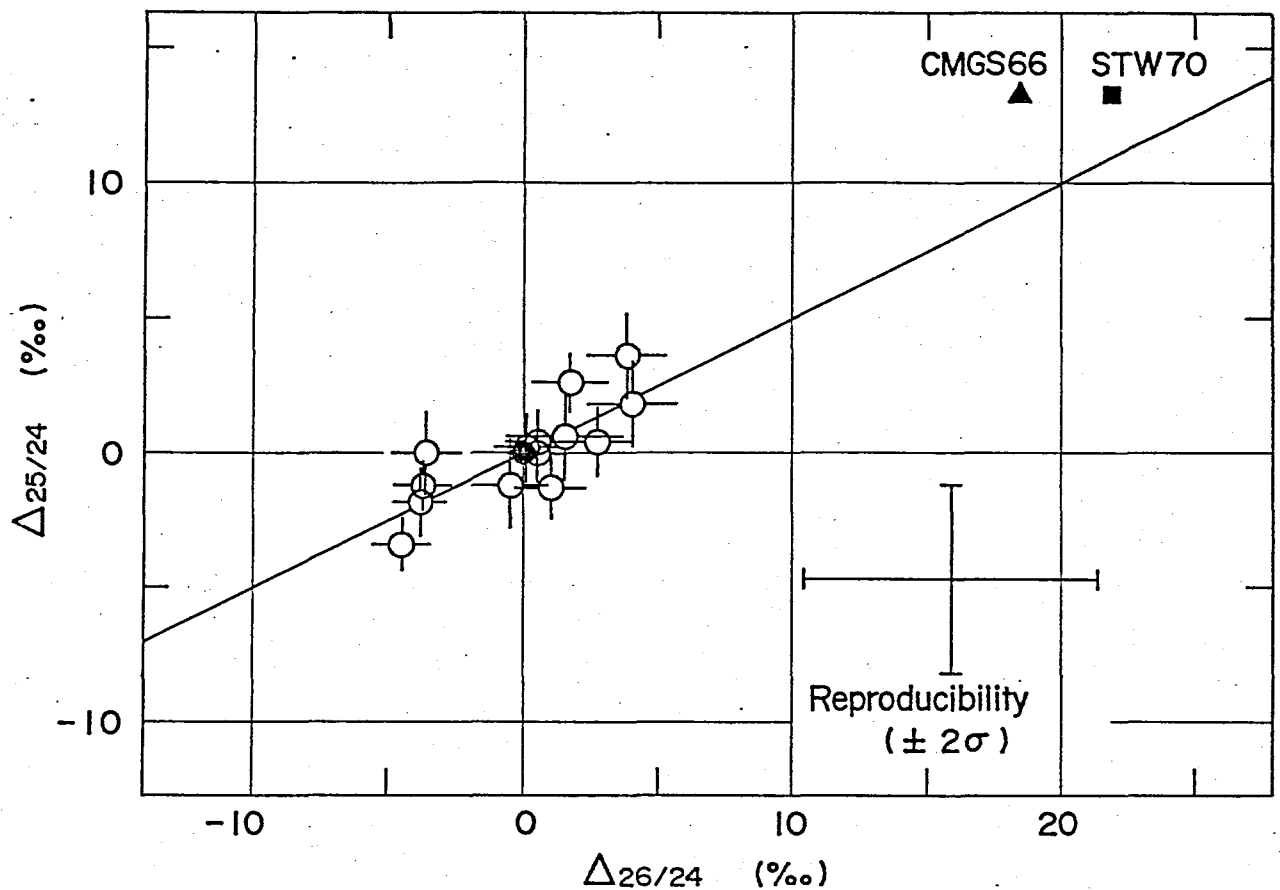


Fig. 4.7 Three isotope plot of the data for the terrestrial forsterite (FO). A straight line with the slope of 1/2 corresponds to the normal mass fractionation line. The absolute abundance ratios reported by Catanzaro et al. (CMGS 66) and Schramm et al. (STW 70) are also marked.

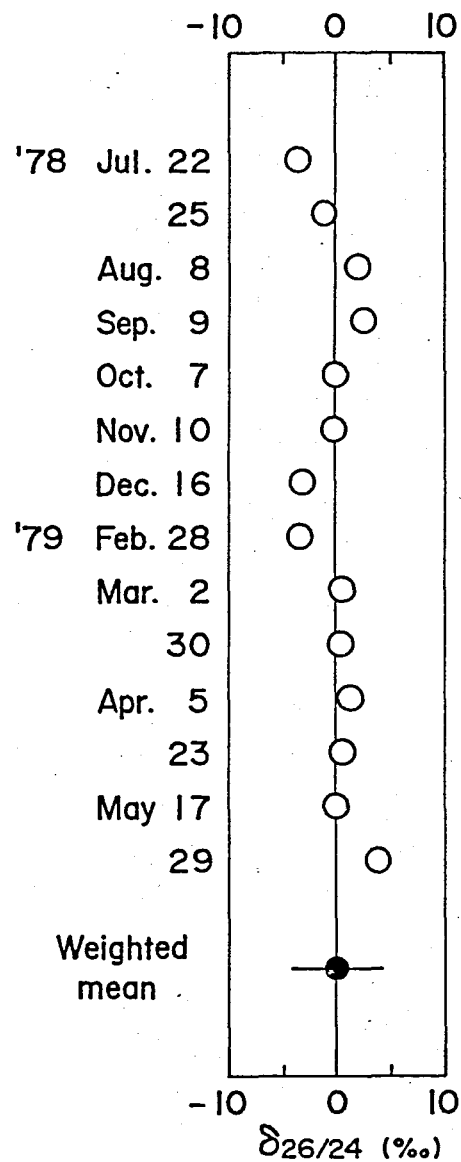


Fig. 4.8 Deviations, $\delta_{26/24}$, after the correction for the normal mass fractionation. These were obtained for the FO sample. The definition of $\delta_{26/24}$ appears in the text. These are plotted in the chronological order. The error bar for the closed circle represents the reproducibility.

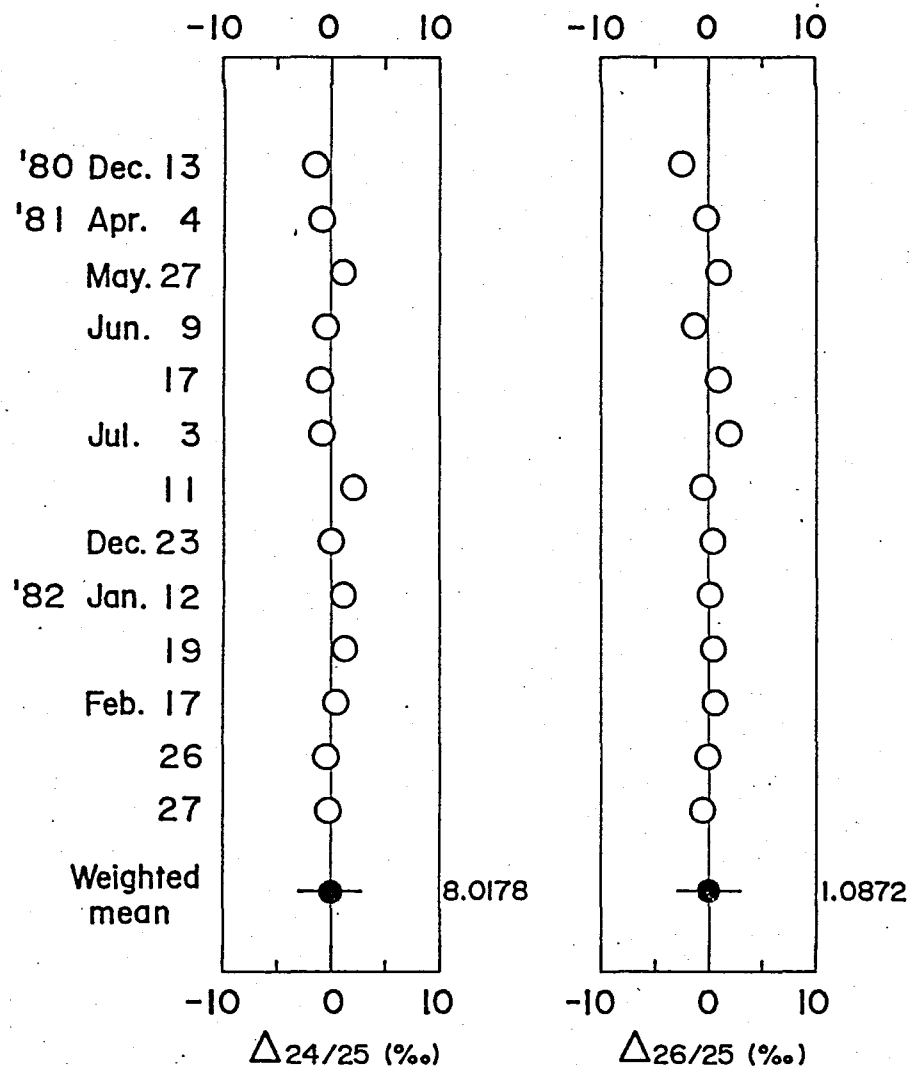


Fig. 4.9 Δ values for the FO sample. These data are plotted in the chronological order. The definition of Δ appears in the text. Values aside the closed circles are the weighted mean values of all these data, and these values were used as reference values in evaluating Δ values. Error bars for the closed circles represent twice the standard deviations.

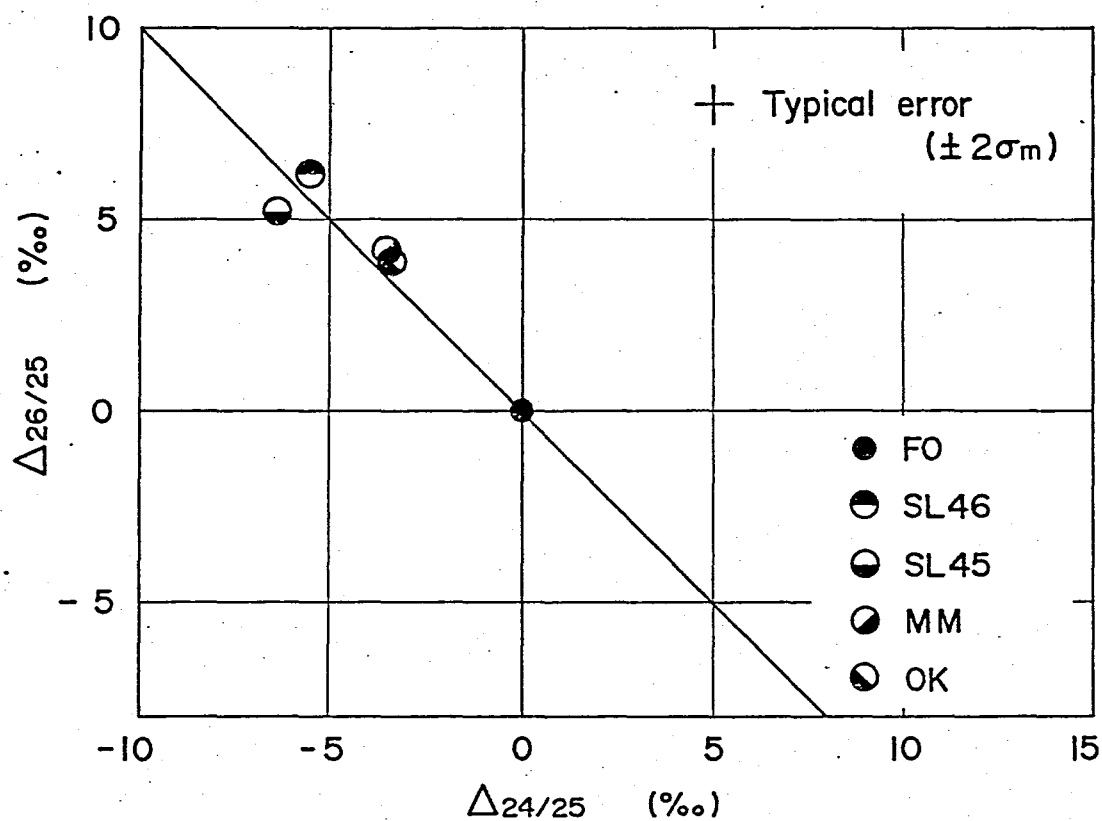


Fig. 4.10 Three isotope plot of magnesium obtained for four terrestrial olivines. These were analyzed as sub-standards. FO: forsterite (Ehime Pref.), SL46: olivine (Hawaii) SL45: olivine (Hawaii), MM: olivine (Antarctica) OK: olivine (Oki Island)

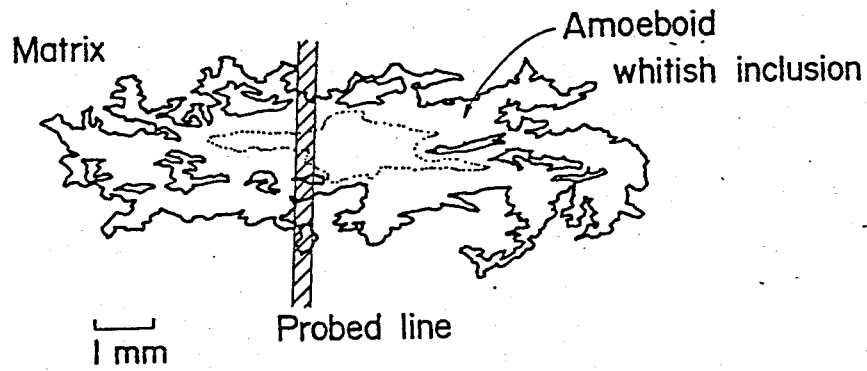


Fig. 4.11 Sketch of the amoeboid whitish inclusion in AL0.

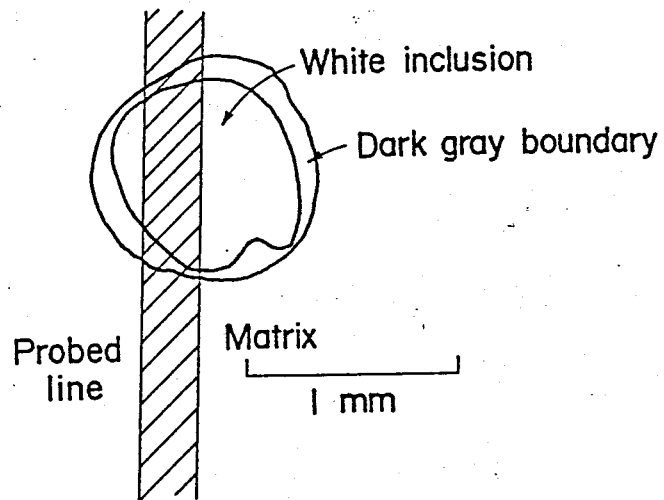


Fig. 4.12 Sketch of the chondrule-like white inclusion in AL1.

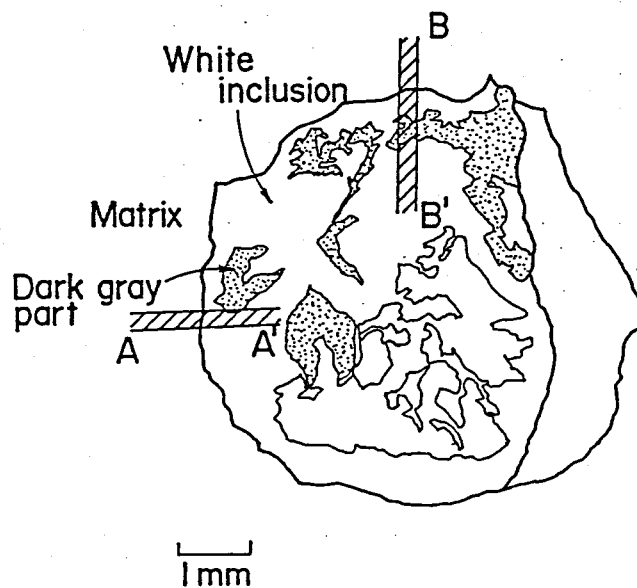


Fig. 4.13 Sketch of the comparatively large white inclusion in AL2.

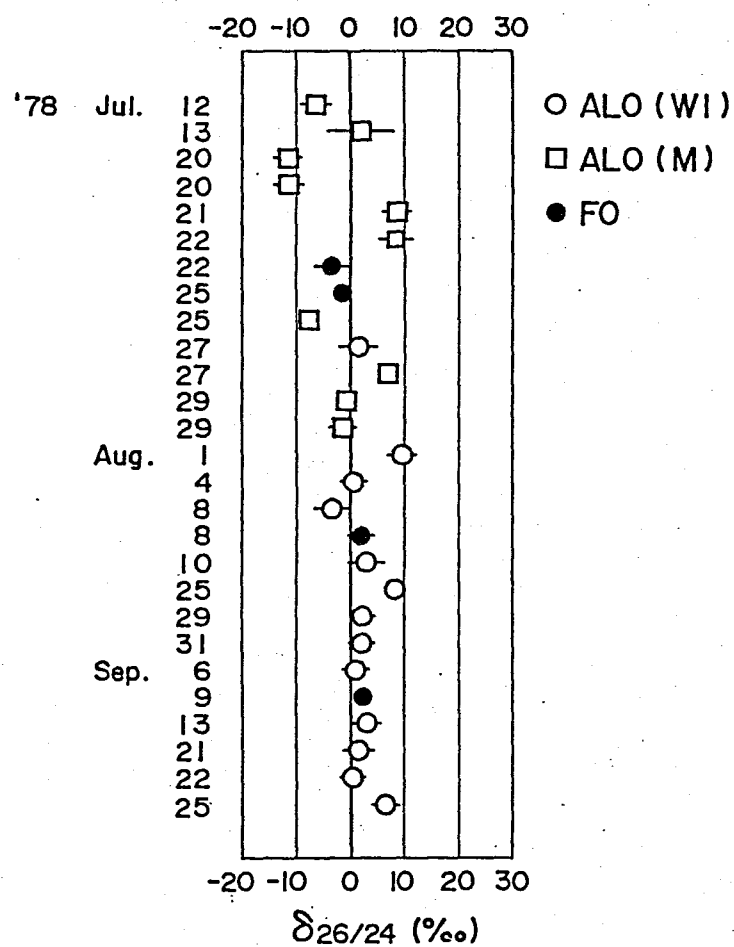


Fig. 5.1 $\delta_{26/24}$ values obtained for AL0 including the amoeboid whitish inclusion. The definition of $\delta_{26/24}$ appears in the text.

AL0(WI): whitish inclusion in AL0

AL0(M): matrix in AL0

FO: terrestrial forsterite (laboratory standard)

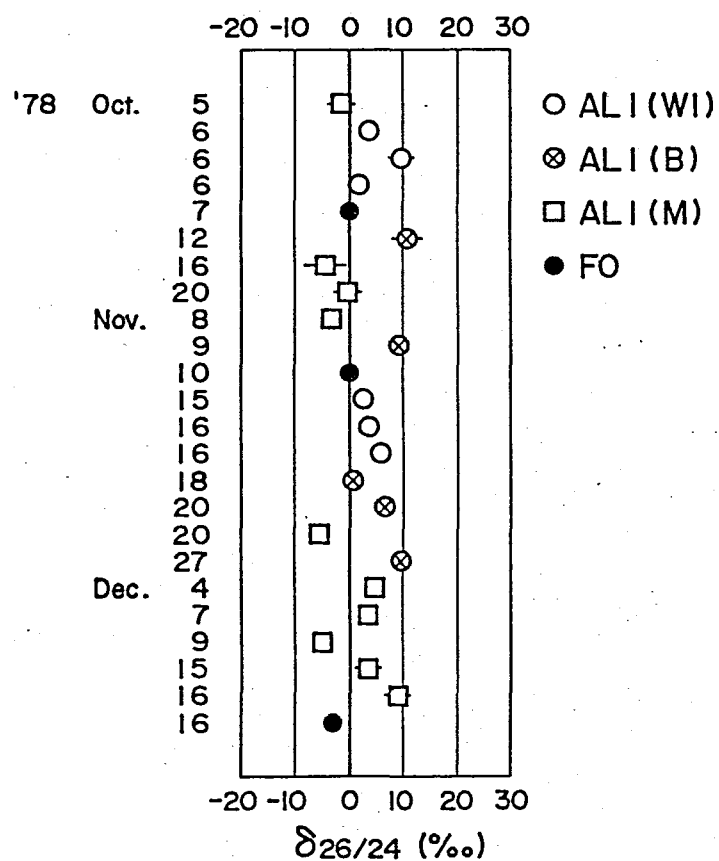


Fig. 5.2 $\delta_{26/24}$ values obtained for AL1 including the chondrule-like white inclusion surrounded by the boundary layer. The definition of the $\delta_{26/24}$ appears in the text.

AL1(WI): white inclusion in AL1

AL1(B): boundary layer surrounding the inclusion in AL1

AL1(M): matrix in AL1

FO: terrestrial forsterite (laboratory standard)

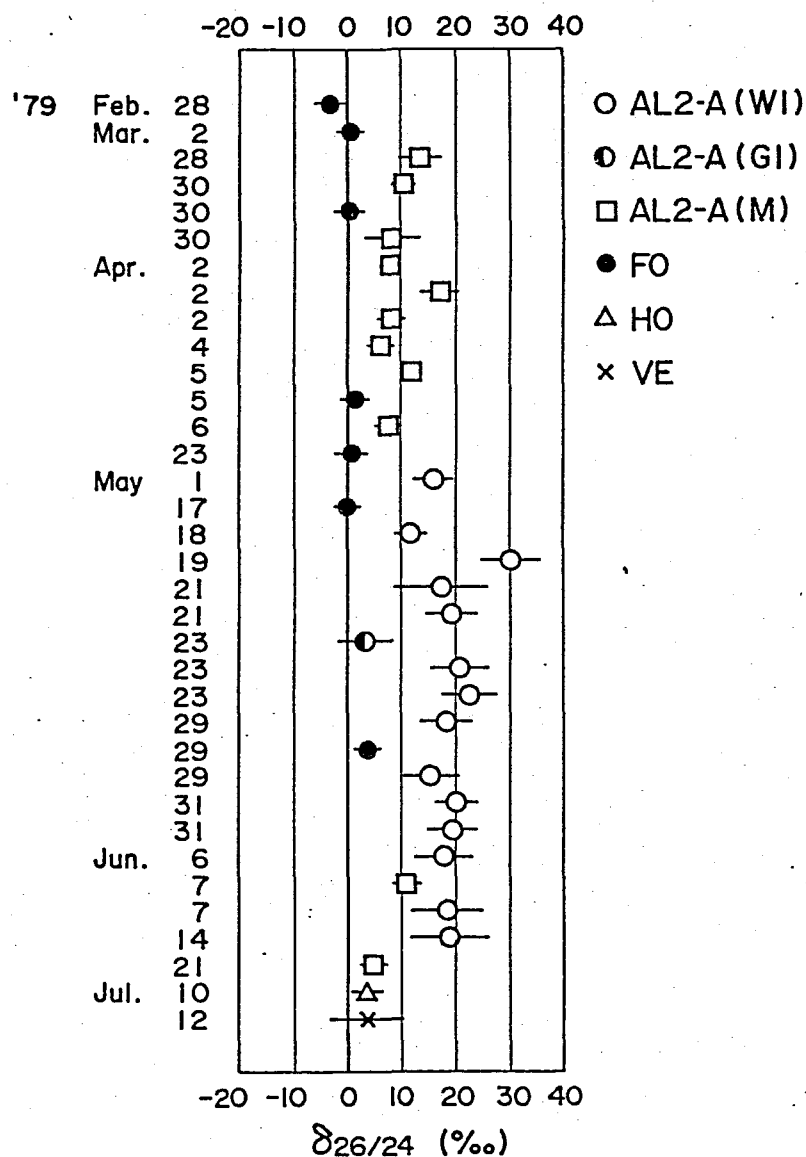


Fig. 5.3 $\delta_{26/24}$ values obtained for AL2 including the large white inclusion. The definition of $\delta_{26/24}$ appears in the text.

AL2-A(WI): white inclusion in AL2 (analysis along AA')

AL2-A(GI): dark gray part in AL2

AL2-A(M): matrix in AL2

FO: terrestrial forsterite (laboratory standard)

HO: terrestrial hornblende, VE: terrestrial vesuvianite

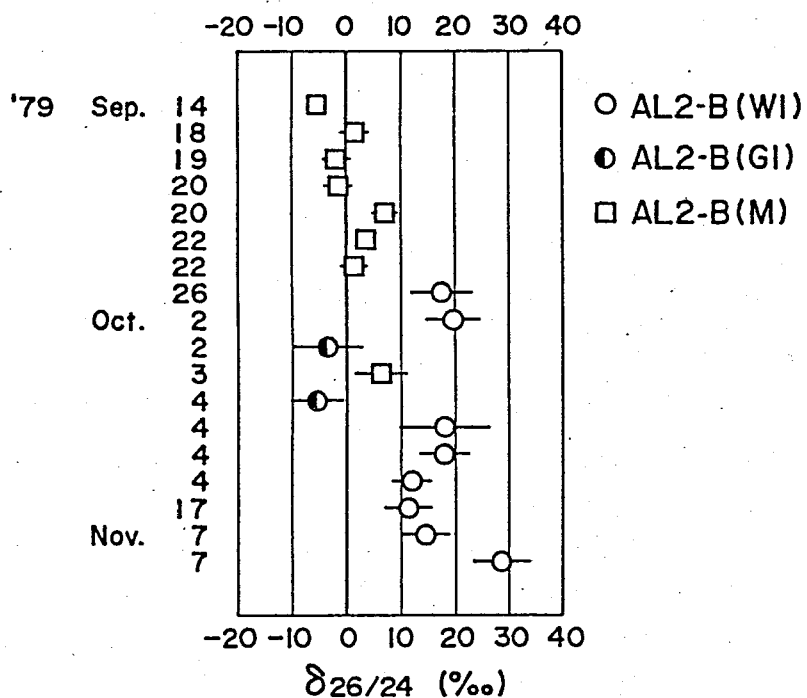


Fig 5.4 $\delta_{26/24}$ values obtained for AL2 including the large white inclusion. The definition of $\delta_{26/24}$ appears in the text.

AL2-B(WI): white inclusion in AL2 (analysis along BB')

AL2-B(GI): dark gray part in AL2

AL2-B(M): matrix in AL2

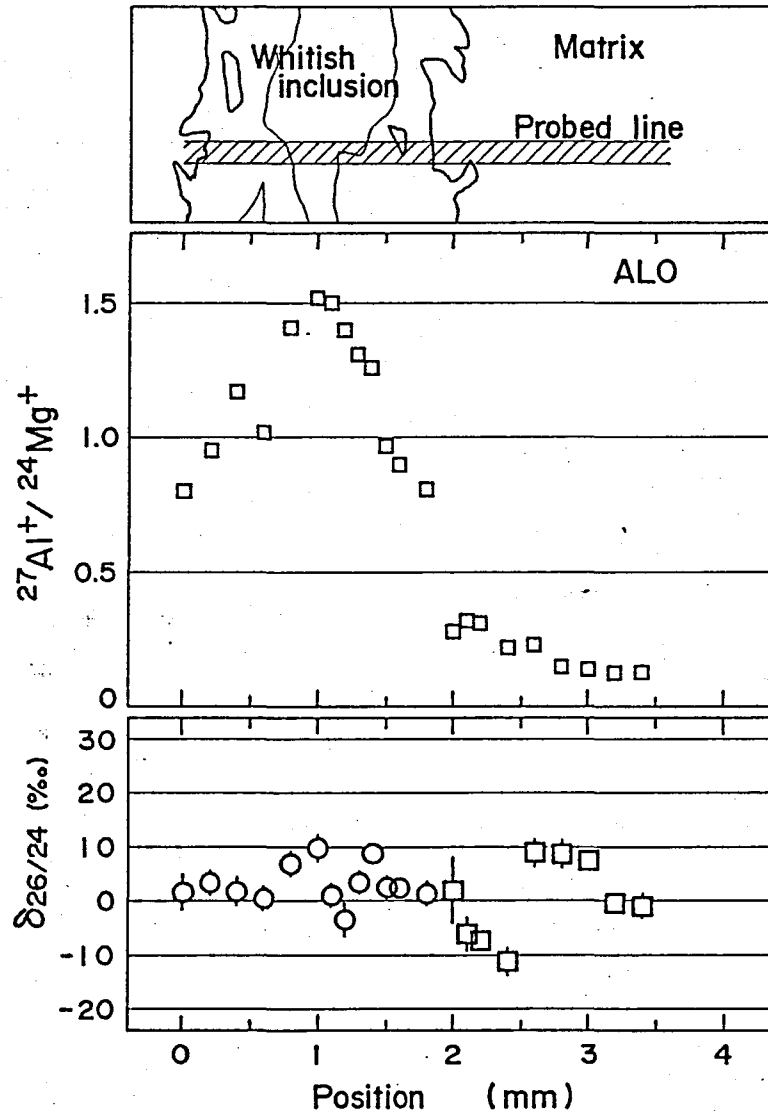


Fig. 5.5 $\delta_{26/24}$ and $^{27}\text{Al}^+ / ^{24}\text{Mg}^+$ as a function of probed position along the probed line for the AL0. Symbols for the plot of $\delta_{26/24}$ are the same as shown in Fig.5.1.

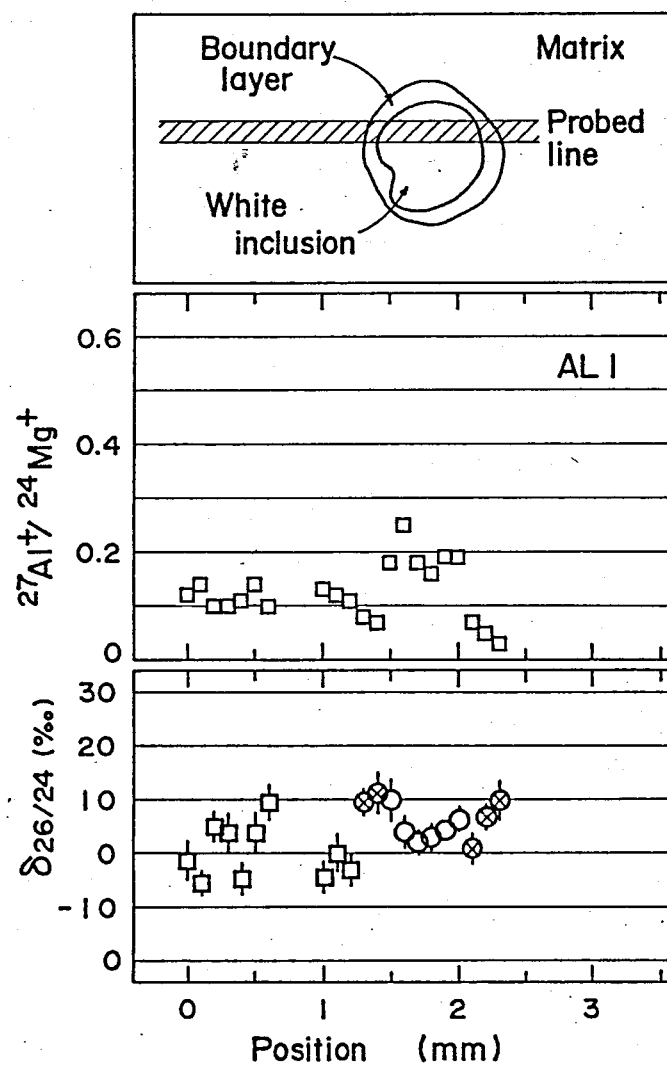


Fig. 5.6 $\delta_{26/24}$ and $^{27}\text{Al}^+ / ^{24}\text{Mg}^+$ as a function of probed position along the probed line for the AL1. Symbols for the plot of $\delta_{26/24}$ are the same as shown in Fig.5.2.

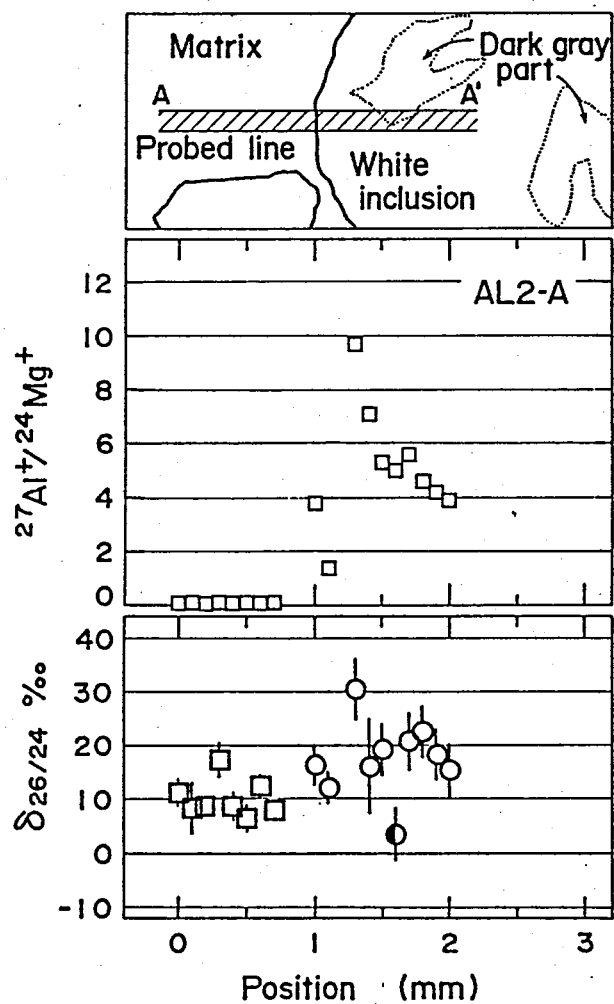


Fig. 5.7 $\delta_{26/24}$ and $^{27}\text{Al}^+ / ^{24}\text{Mg}^+$ as a function of probed position along the probed line AA' for the AL2. Symbols for the plot of $\delta_{26/24}$ are the same as shown in Fig. 5.3.

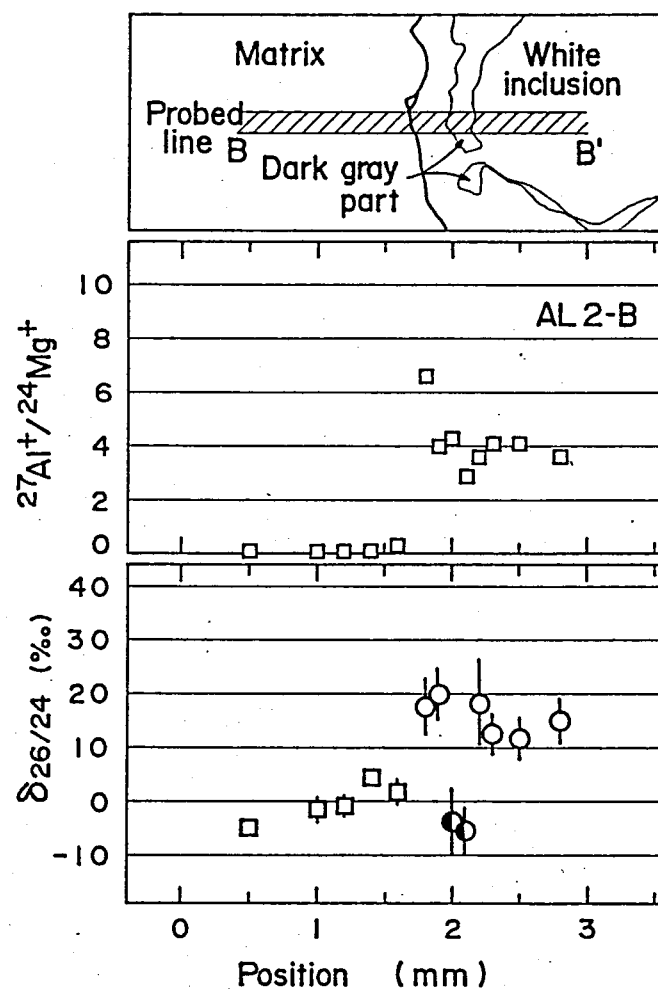


Fig. 5.8 $\delta_{26/24}$ and $^{27}\text{Al}^+ / ^{24}\text{Mg}^+$ as a function of probed position along the probed line BB' for the AL2. Symbols for the plot of $\delta_{26/24}$ are the same as shown in Fig.5.4.

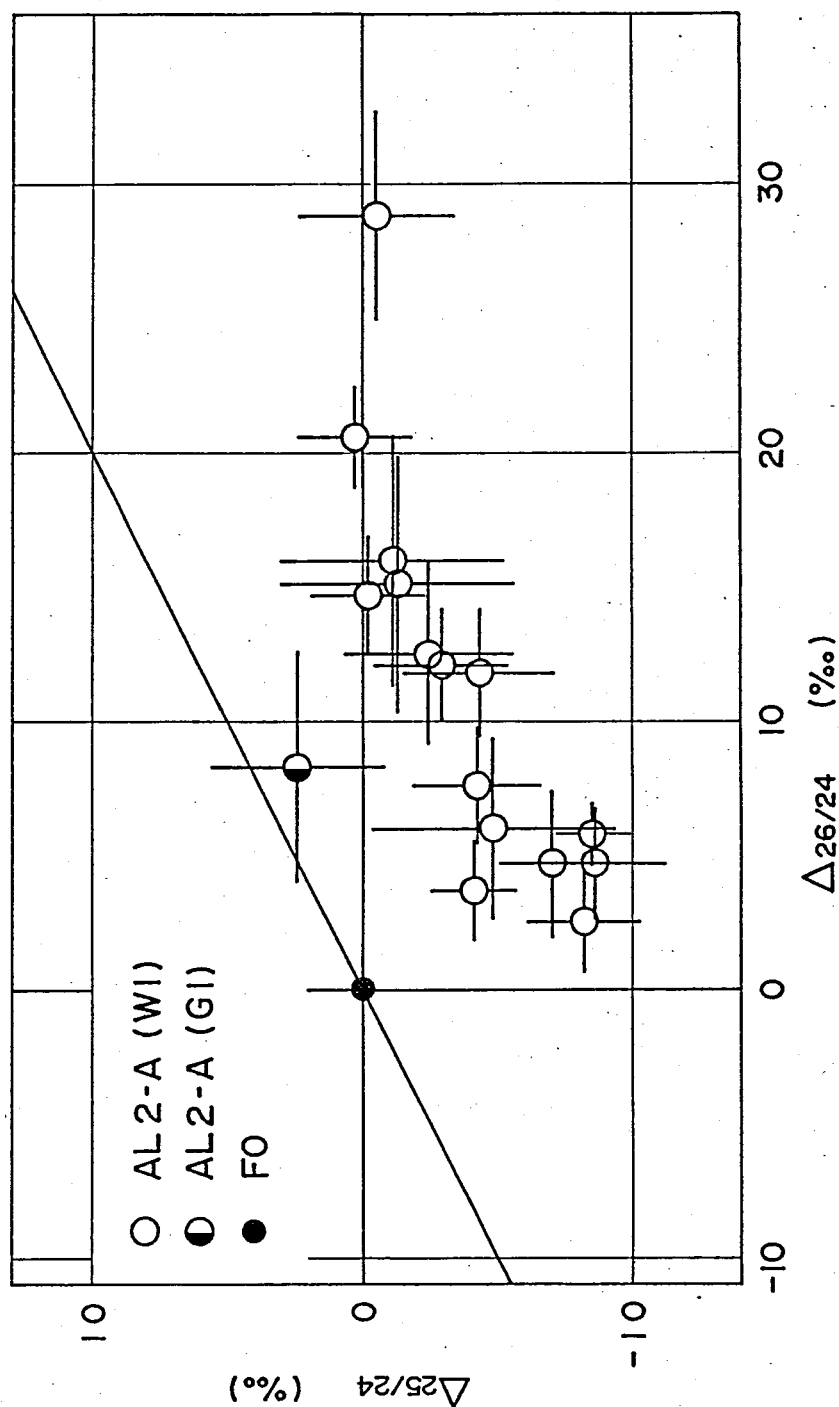


Fig. 5.9 Three isotope plot of magnesium for the white inclusion in the AL2. These data were obtained along the probed line AA'. A straight line with the slope of 1/2 represents the normal mass fractionation line.

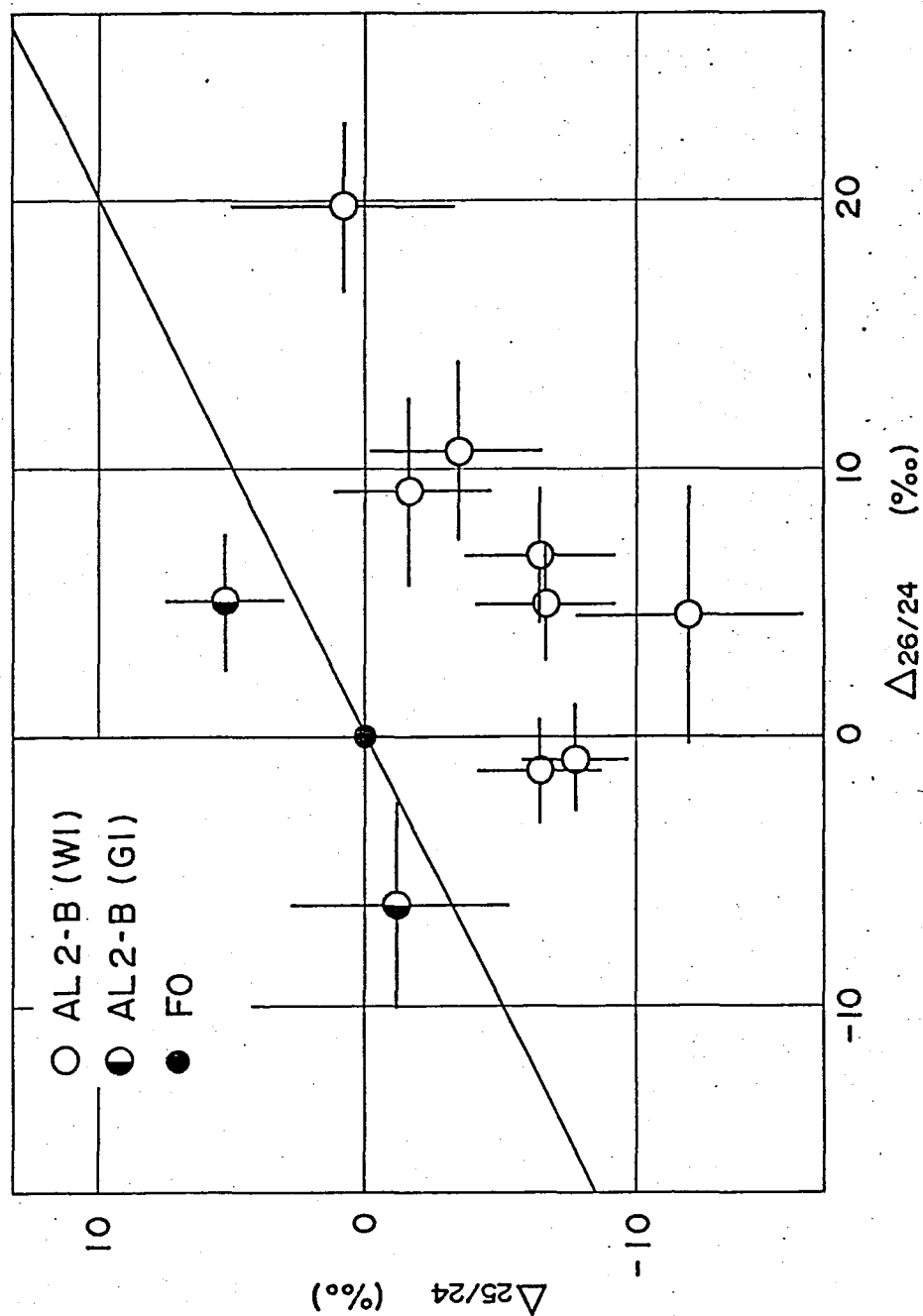


Fig. 5.10 Three isotope plot of magnesium for the white inclusion in the AL2. These data were obtained along the probed line BB'. A straight line with the slope of 1/2 represents the normal mass fractionation line.

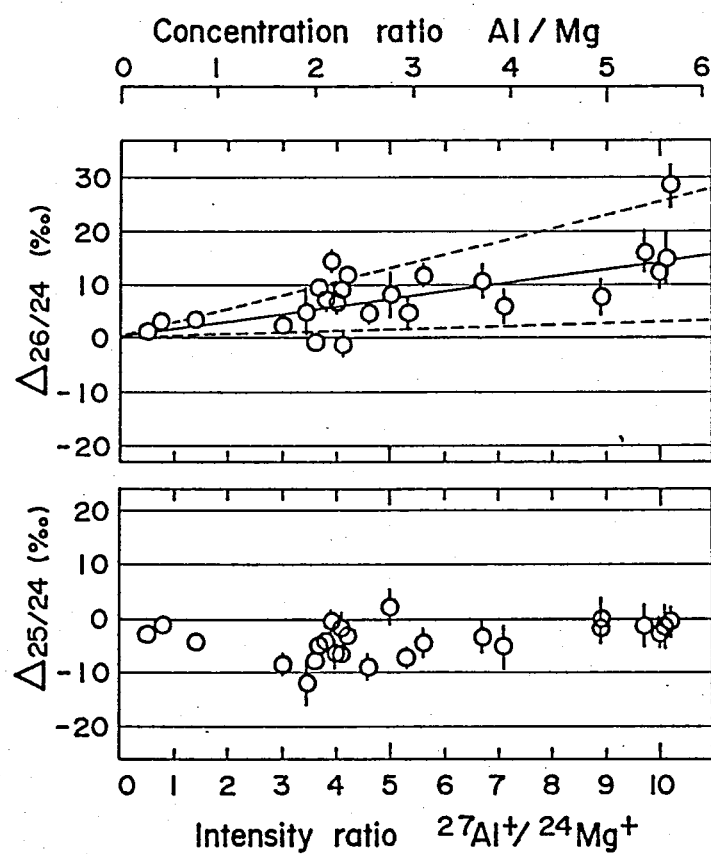


Fig. 5.11 $\Delta_{26/24}$ and $\Delta_{25/24}$ versus Al/Mg ratio showing the correlation between $\Delta_{26/24}$ and Al/Mg. These data are the same as plotted in both Figs. 5.9 and 5.10.

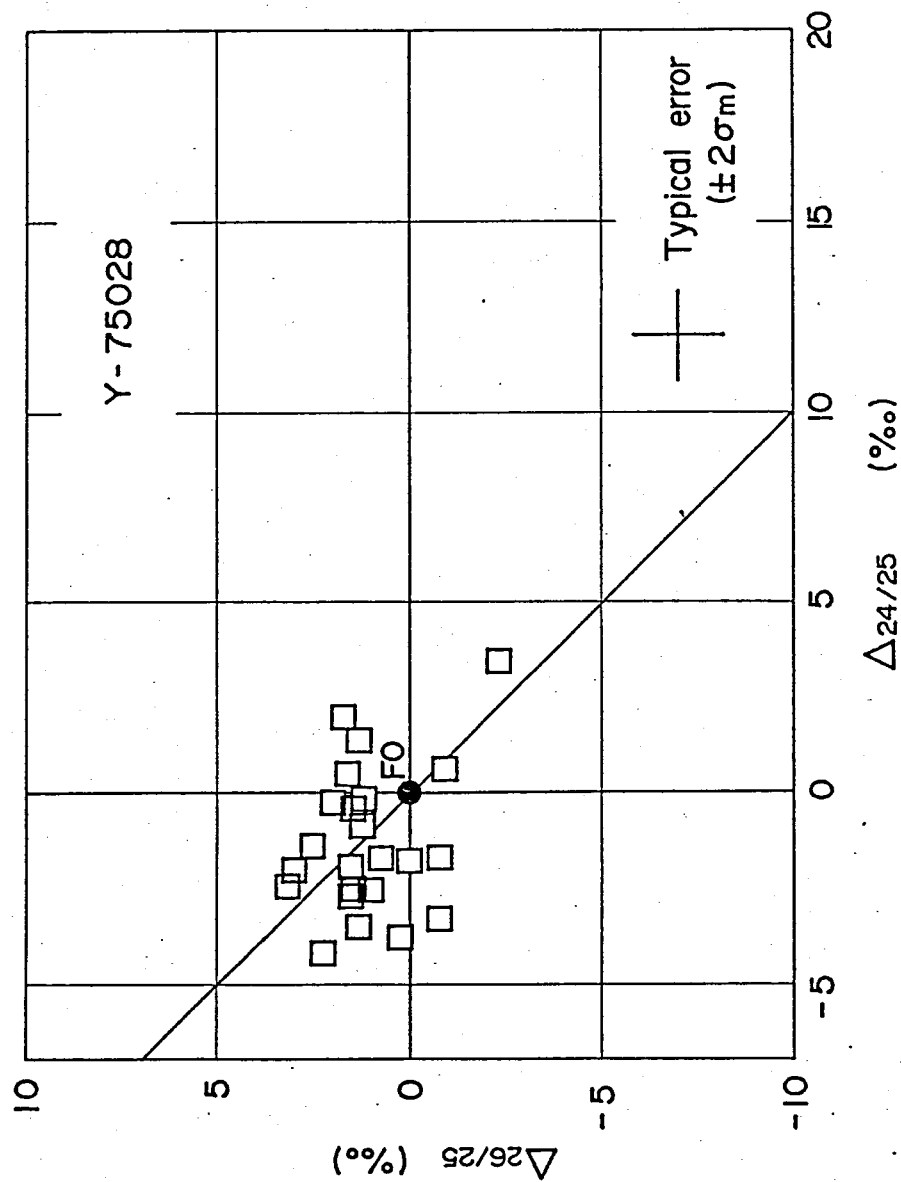


Fig. 5.12 Three isotope plot of magnesium for the matrix portions of Y-75028. The plotted data were obtained for the portions where the concentration ratio of Al/Mg is lower than 0.13.

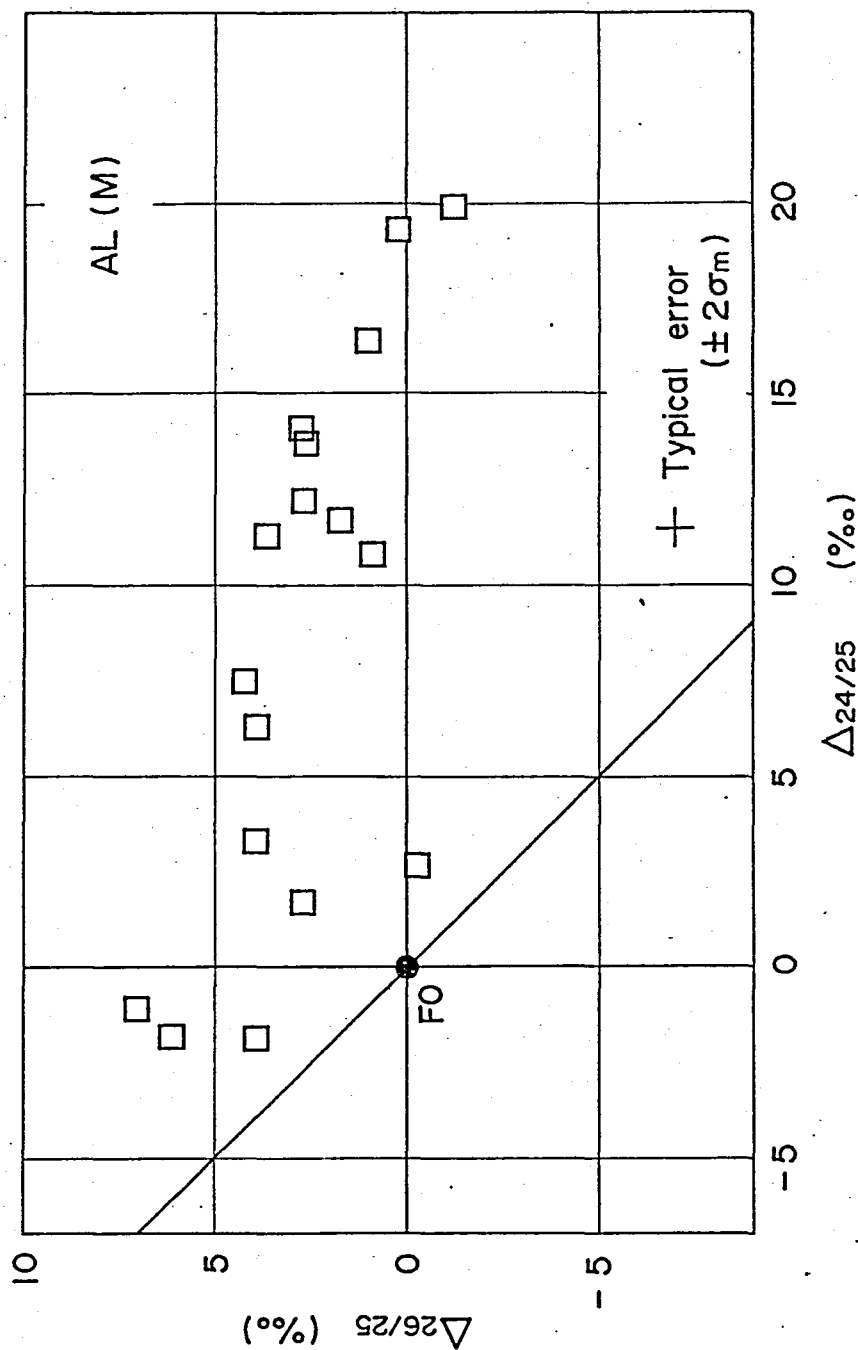


Fig. 5.13 Three isotope plot of magnesium for the matrix portions of Allende. The data were obtained for the portions where the concentration ratio of Al/Mg is less than 0.13.

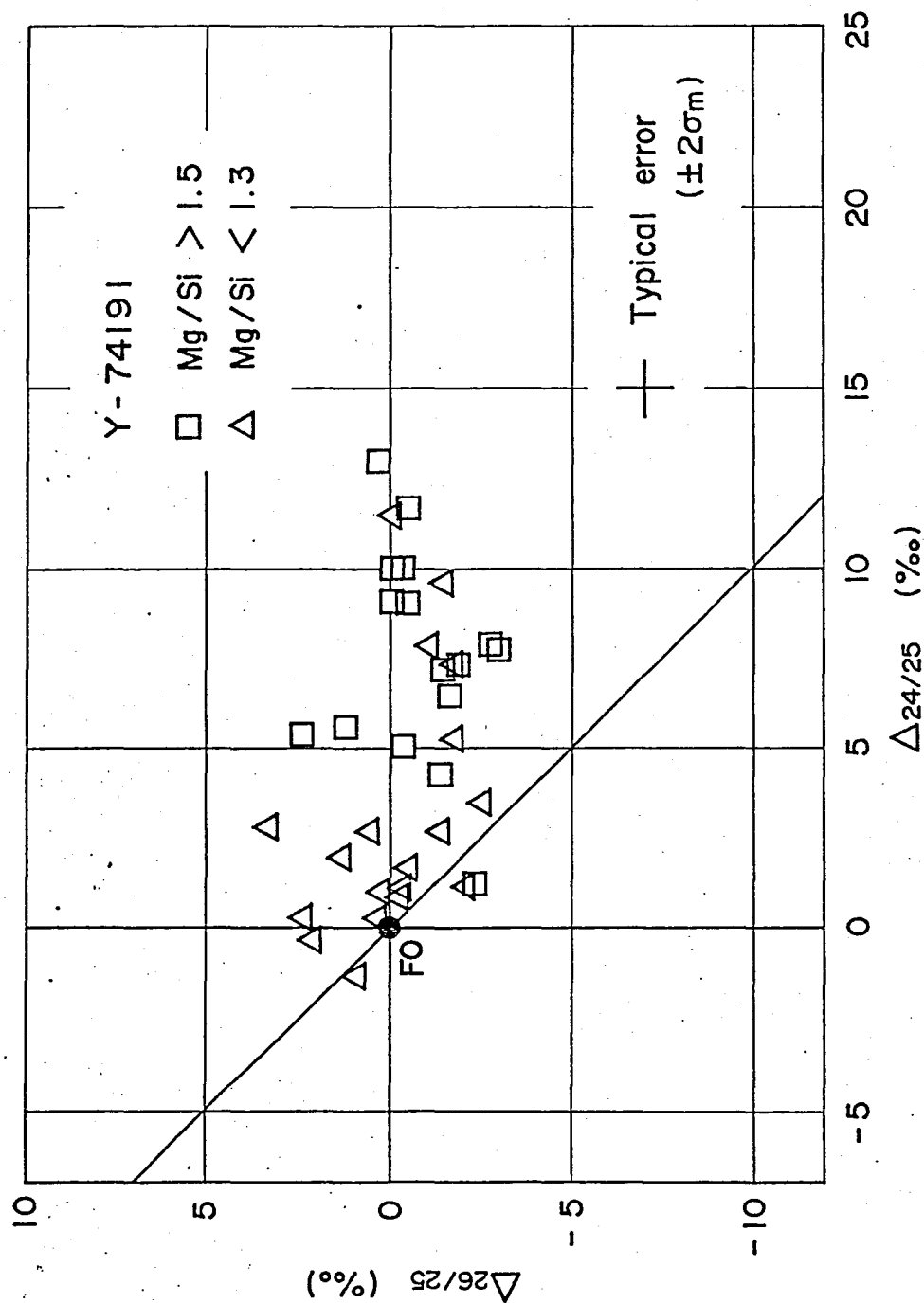


Fig. 5.14 Three isotope plot of magnesium for the matrix portions of Y-74191. The data were obtained for the portions where the concentration ratio of Al/Mg is less than 0.13.

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