Determination of Rb-Sr age for separated chondrules from the Allegan chondrite

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Rb-Sr isotopic study as well as petrologic investigations have been done on sixteen individual chondrule samples separated from the Allegan (H5) chondrite. The Rb-Sr chondrule isochron does not define a very precise age (4.42±0.17 Ga) and $^{87}Sr/^{86}Sr$ initial ratio (0.6997±0.0014) due to the very small variation in Rb/Sr ratio for the chondrules, whereas the model ages give a much more specific value (4.52±0.02Ga). These ages may probably be indicative of the time of primary process(es) (e.g. chondrule formation) rather than secondary (metamorphic reheating) or tertiary (shock heating) processes in and/or on the H-chondrite parent body (or bodies). Chemical compositions of olivines (Fa=17.5mol%, mean deviation (MD)=1.3%) and orthopyroxenes (Fs=15.4mol%, MD=3.2%) are moderately homogenized among the chondrules, while certain chemically unequilibrated features are also distinctly preserved in this chondrite. Homogeneity of the mineral compositions, therefore, could not have been caused by thermal metamorphism but were probably inherited from processes prior to aggregation of the components. The overall implications of the chondrule isochron and model ages in relation to the petrological features and in comparison with Rb-Sr data of other chondrites of different chemical groups and petrologic types are discussed.

Keywords: chondrule, age, Rb-Sr isochron, Allegan, H- chondrite, meteorite, metamorphism, olivine, pyroxene

I. Introduction

Chondrites are most common among meteorites and are considered to be the most primitive chemically. As to their physical structure, chondrites consist of various mechanically-assembled components of different nature and origin. Chondrules are structurally conspicuous constituents of chondrites and are believed to be the quenched droplets of precursory materials once melted in the early stage of the solar system (e.g. Hewins et al., 1981). The origin of chondrules, therefore, has been a topic of extensive study and as much controversy among reseachers on the early processes in the solar system. Age investigations of chondrites are indispensable for adequate understanding of solar processes. Extensive efforts have therefore been focused on the subject, mainly utilizing long-life radionuclide-daughter pairs. Most of the investigations have been done by the K-Ar method. This method of dating, however, may not be suitable for determining the age of primary processes in the

evolutionary history of meteorites because of ease of Ar release by later processes (e.g. shock processes). For a better approach to the purpose, determinations of isochron ages by Rb-Sr, Sm-Nd, and U-Th-Pb methods have been widely used. Among these, the Rb-Sr method has advantage over the others because of higher abundances in Rb and Sr and variability of Rb/Sr ratio in chondritic materials. Furthermore, recent developments in dating techniques have made it possible for extremely small (≤1mg) samples of chondritic materials to be dated. Attempts to determine the Rb-Sr ages of chondrules have so far been made on several chondrites (Murthy and Compston, 1965; Shields et al., 1966; Grey et al., 1973; Tatsumoto et al., 1976; Hamilton et al., 1979; Yoshikawa and Nakamura, 1992; Evensen et al., 1979; Minster and Allègre, 1979, 1981; Papanastassiou and Wasserburg, 1969, 1981). The implications of the chondrule ages, however, have not been fully understood. This paper documents our preliminary study on ordinary chondrites, demonstrating a precise determination

of Rb-Sr age for chondrules separated from the Allegan chondrite (H5) on which petrologic and chemical features have been relatively well investigated.

II. Sample and experimental procedures

1. Sample preparation

The Allegan chondrite specimen used in this study was provided by Dr. N. Nakamura of the Department of Earth Sciences, Kobe University. A detailed study on major and trace element (K, Rb, Sr, Ba, and rare earths) abundances (Nishikawa, 1984) and a preliminary investigation on the Rb-Sr systematics (Nishikawa et al., 1990) have so far been made on chondrules separated from a part of the specimen. Sixteen chondrules (2.8 to 32.2mg) were taken out from this same specimen for the study. After observation of the separated chondrules under a binocular microscope, each chondrule was crushed into a few fragments in an agate mortar. Subsequently, a small fragment was picked out from each chondrule to make polished thin sections for X-ray microprobe analyses whilst the remaining portion was weighed for Rb-Sr analysis.

Two to twenty milligrams (2-20mg) each of the samples was first weighed and put in teflon vials. These were then decomposed with 0.2-0.6 ml of a 1:1 mixture of $HF-HClO_4$, depending on the sample weights (approx. 1mg for 0.1ml). The vials were sealed and left at room temperature for several days after which they were heated overnight on a hot plate at $\sim150°C$. After complete decomposition, they were unsealed and the contents evaporated to dryness on a hot plate in a clean bench. To replace fluoride with chloride, twenty drops of 6N HCl were subsequently added to the dried samples and the vials resealed for further decomposition on the hot plate. The contents were evaporated to dryness at $\sim200°C$ to get rid of perchloric acid completely. The next step involved addition of 0.5 ml of 2N HCl to the dried samples, followed by 0.5 ml of double distilled water. The resultant solutions were split into two, 4/5 and 1/5, for the determinations of Sr isotopic composition and precise Rb and Sr concentrations, respectively. Except for chondrule sample Nos. 1 and 9 where 0.42ml of resin (Dowex AG50W-12X, 200-400 meshes) was taken, 0.26ml of the same resin was employed in the cation-exchange chromatography in H^+ form to separate Sr in the

Table 1. Procedural blanks for Rb-Sr analyses of the Allegan samples. Amounts are shown in 10^{-9} g.

	A	B
Rb	3	10
Sr	23	29
$^{87}Sr/^{86}Sr$	46	60

solutions meant for Sr isotope determination. To the other solutions (for Rb-Sr determinations by the isotope dilution method), a few drops of ^{87}Rb-^{84}Sr composite spike solution were added and weighed. The spiked solutions, in vials, were evaporated to dryness on a hot plate. Further, a few drops of 6N HCl were added to the dried samples and again re-evaporated to dryness. This operation was done to homogenize Rb-Sr isotopic composition of the solutions. These fractions (i.e. Rb-Sr fractions) were not subjected to Rb-Sr separation but to Rb-Sr analyses by direct-loading mass spectrometry (Nakamura et al., 1989). Blank specimens for the two determinations (Sr isotope and Rb-Sr) were similarly prepared using the above-mentioned procedures but without samples. Blank levels estimated for the above whole procedures are shown in Table 1.

2. Mass Spectrometry

The isotopic measurements for Rb-Sr systematics were carried out at the Institute for Study of the Earth's Interior, Okayama University, using Finnigan MAT261 thermal ion-ization mass spectrometer. We carried out the Sr isotopic measurements by using tungsten-single filaments and Ta-activator solution. The procedures employed in this study are basically similar to those in previous works (e.g. Birck and Allègre, 1978; Okano et al., 1984, Nishikawa et al., 1990; Yoshikawa et al., 1992). The filament current for the Sr isotopic measurement ranged from 3.38A to 3.60A, giving a stable ^{88}Sr signal of $1.5-3x10^{-11}A$. Isotopic measure-ments for the NBS987 standard (Sr 50ng) were also made in the course of this study ($^{87}Sr/^{86}Sr=0.710279\pm 0.000018$; n=3).

In the Rb and Sr concentration determination, Ta-Re double filaments were used. Each sample was loaded directly onto a Ta-filament without separating Rb and Sr from the major elements. Ta- and Re-filament currents used to measure Rb concentration ranged from 0.3 to 0.5A and 1.8 to 2.0A, respectively. In the subsequent determi-

nation of Sr concentrations using the same filaments as above, the current ranged from 2.1 to 2.4A and 3.2 to 3.4A, respectively. Measurements of the blank specimens were carried out at Kobe University, using JEOL JMS-05RB mass spectrometer equipped with a secondary electron multiplier and correspondingly same procedures as outlined above.

3. X-ray microprobe analyses

From the sixteen chondrule fragments earlier picked out for microprobe analyses, two polished thin sections were made. Mg, Fe, and Ca elemental compositions in olivines and pyroxenes, for each chondrule, were determined with JEOL JXA-733 X-ray microprobe analyzer operated at 15kV accelerating voltage and 2×10^{-8}A specimen current measured on Cu metal. This facility is available at the Okayama University. The correction procedure was that of Bence and Albee (1968). In some samples, accurate data may not have been obtained because the diameter of beam was large compared to mineral grain sizes.

III. Results

1. Chondrule textures

The chondrules studied are divided into four textural types (Fig. 1 and Table 2). Among the sixteen chondrules, six (i.e. Nos. 1, 4, 5, 9, 15, 16)

are identified as radial pyroxene chondrules (RP). The latter consist essentially of bunches of Ca-poor pyroxene needles (apparent width 5 to 120μm). The apparent widths of pyroxene needles vary from chondrule to chondrule and even at different locations within a single chondrule. Four chondrules (Nos. 2, 8, 11, 14) are porphyritic olivine chondrules (PO) consisting mainly of subhedral olivine crystals (up to 400μm long) set in matrices of dark, fine aggregates. A compound texture (RP/PO) is also observed in three chondrules (Nos. 10, 12, 13), where porphyritic olivines are locally set in dominant radial-pyroxene areas. Chondrule Nos. 6 and 7 are microcrystalline chondrules (MC); No. 6 consists of Ca-poor pyroxenes of submicron to several micron size and No. 7 several-micron grains of both olivines and Ca-poor pyroxenes. Chondrule No. 3 was observed to be a PO chondrule from a visual inspection but the details were not clarified due to sample loss in the course of thin section preparation.

2. Chemical compositions in olivines and pyroxenes

Chemical compositions (Fe-Mg-Ca) were determined for olivines from nine chondrules and for pyroxenes from thirteen chondrules (Table 2 and Fig. 2). Regardless of chondrule texture, size, and shape of grains, Fa contents in olivines are fairly constant (mean Fa content = 17.5 mol%,

Table 2. Mineral compositions and Rb and Sr abundances in the Allegan chondrules.

Chondrule	Texture	Weight (mg)	Fe/(Fe+Mg) (mol%) olivine	Fe/(Fe+Mg) (mol%) opx	Rb (ppm)	Sr (ppm)	Rb/Sr
1	RP	32.2	-	15.48 (7)	2.22	10.2	0.218
2	PO	6.1	17.61 (7)	15.81 (1)	2.35	12.8	0.184
3	PO ?	5.7	-	-	2.63	14.1	0.187
4	RP	7.9	-	15.71 (7)	2.05	11.8	0.174
5	RP	5.6	-	15.75 (7)	1.94	10.7	0.181
6	MC	6.0	-	16.75 (8)	1.36	9.1	0.149
7	MC	3.7	18.06 (1)	17.49 (1)	2.18	13.1	0.166
8	PO	3.9	17.57 (5)	15.59 (1)	2.91	15.3	0.190
9	RP	10.6	17.57 (2)	15.48 (7)	2.56	12.5	0.205
10	RP/PO	3.3	17.50 (2)	15.10 (3)	1.99	12.6	0.159
11	PO	6.4	17.45 (8)	15.03 (1)	2.50	14.1	0.178
12	RP/PO	2.8	17.16 (2)	15.10 (6)	2.38	11.9	0.200
13	RP/PO	4.0	17.11 (3)	15.12 (4)	2.83	14.7	0.193
14	PO	3.8	17.63 (6)	-	4.35	24.1	0.181
15	RP	3.6	-	15.44 (4)	2.29	11.7	0.195
16	RP	3.7	-	-	2.06	12.2	0.169

RP, PO, and MC represent radial pyroxene, porphyritic olivine, and microcrystalline chondrules, respectively. Figures in parentheses indicate number of points analyzed.
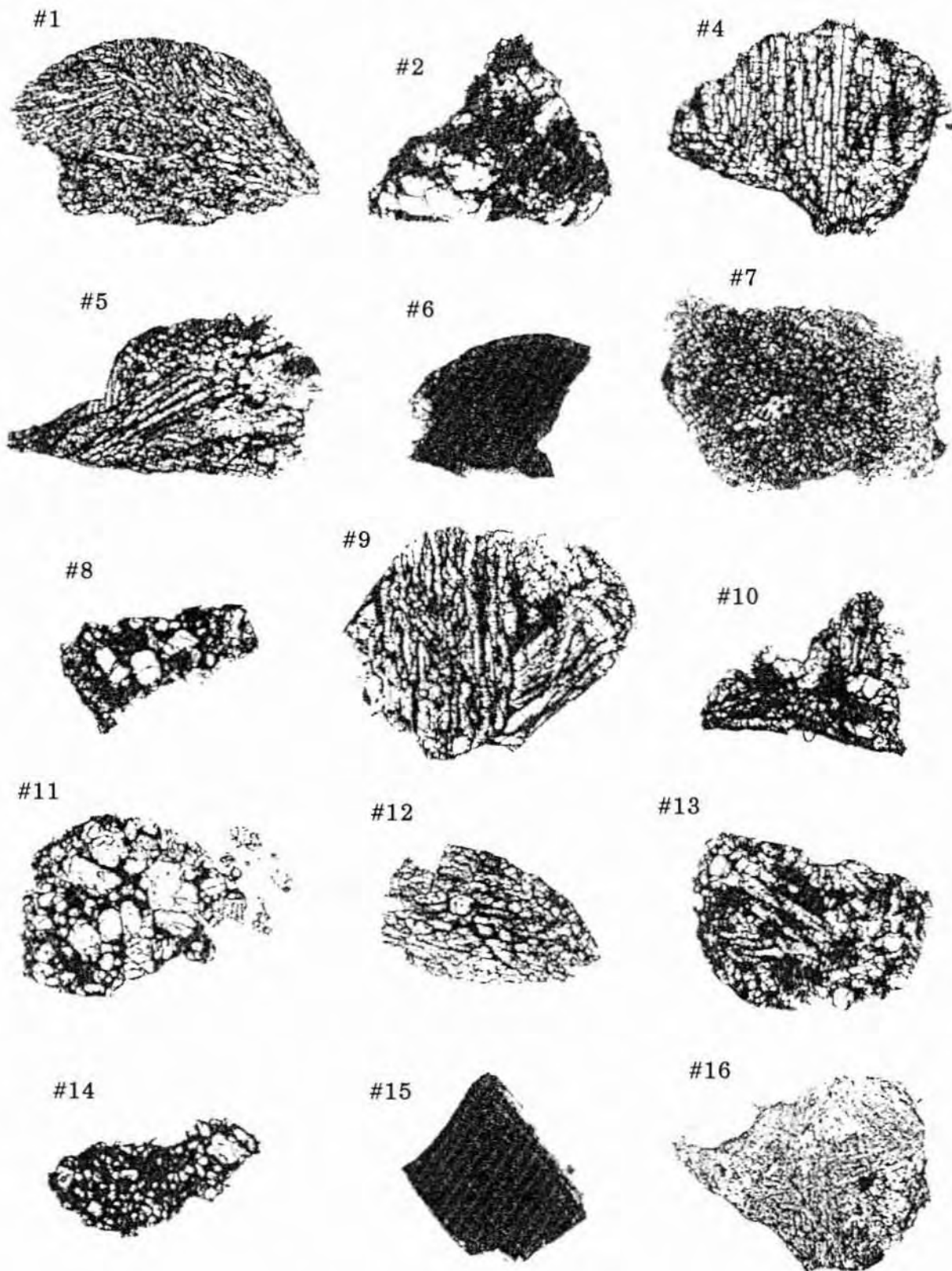
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Fig. 1 Photomicrographs of textures of chondrules from the Allegan chondrite. Magnification is x 30 in all cases except for chondrule #1 (x 15). Descriptions are given in the text.

MD = 1.3%). The mean value is quite typical for an "equilibrated" H-chondrite (e.g. Dodd, 1981) but the degree of homogeneity is comparable to that of type 4 chondrite rather than type 5. Furthermore, the CaO abundances in the olivines (from 0.03 to 0.70 wt.% with a mean value = 0.09 wt.%) are higher than those of well "equilibrated" ones (0.02-0.05 wt.% for types 5 and 6; Dodd, 1981), supporting the above view that this chondrite might possibly be classified as type 4 instead of type 5. The chemical compositions in orthopyroxenes determined for the thirteen chondrules (mean Fs content = 15.4 mol%; MD = 3.0%) are also within the range for "equilibrated" H-chondrites (Fig. 2b) but are somewhat variable compared to olivines. Those from microcrystalline chondrules (Nos. 6 and 7) tend to be slightly more Fe-rich (Table 2). The mean composition of olivines and orthopyroxenes reported above are, however, in good agreement with those by Keil and Fredriksson (1964) (Fa in olivine = 17.5 mol%, Fs in orthopyroxene = 16.0 mol%). As clino-pyroxene was rarely found among the fifteen chondrules examined, chemical analysis was made on only two grains found in Nos. 5 and 15 chondrules. The chemical compositions indicate that they are of endiopsidic ($En_{57.3}Fs_{10.4}Wo_{32.3}$) and magnesium pigeonitic ($En_{76.1}Fs_{13.1}Wo_{10.8}$) compositions respectively (Fig. 3), both of which are normally not involved in "equilibrated" chondrites.

3. Rb-Sr systematics

PO chondrules tend to have relatively higher Rb and Sr contents but similar Rb/Sr ratios (0.178-0.190) which are close to whole rock values (0.178 and 0.188) reported by Nishikawa et al. (1990) and Minster and Allègre (1979). This might be explained by the fact that PO chondrules have more interstitial fine-grained materials, and Rb and Sr abundances are controlled mainly by amounts of such components because incompatible elements including Rb and Sr tend to concentrate in interstitial materials of chondrites. A microcrystalline orthopyroxene chondrule (No. 6) has the lowest values of Rb/Sr ratio and Rb and Sr contents among the sixteen chondrules (Table 2), while radial orthopyroxene chondrules show rather higher Rb/Sr ratio with common abundance values of both elements. The remaining ones are intermediate in both elemental abundances and ratio. The overall
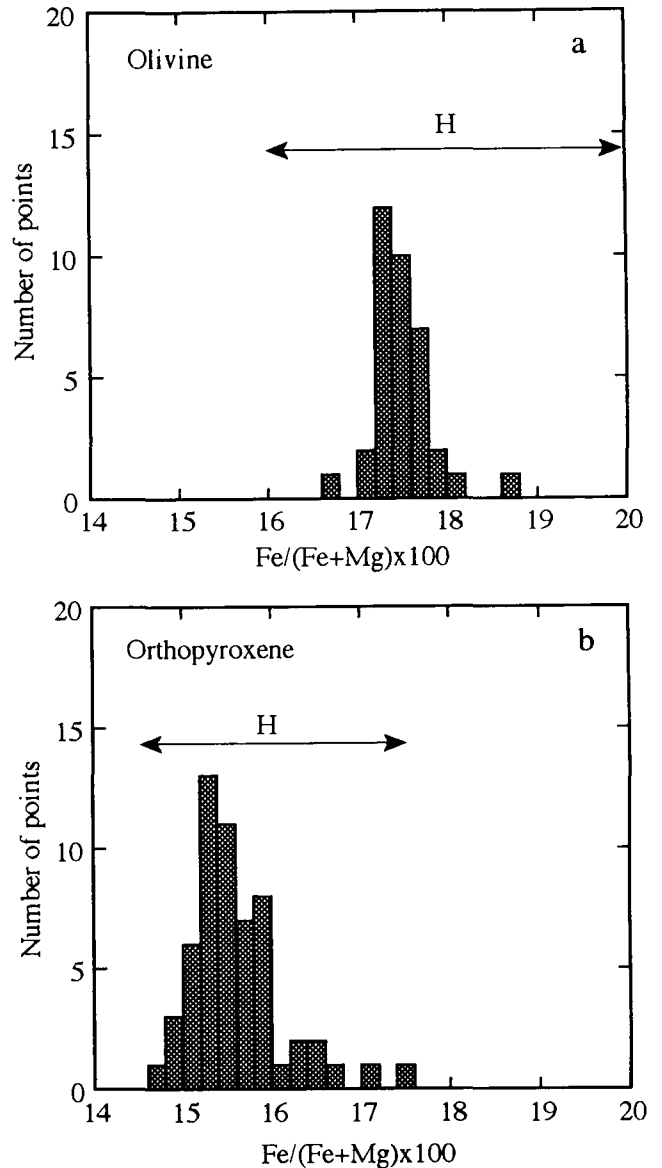

Fig. 2. Frequency diagrams of Fe/(Fe+Mg) ratios in olivines (a) and orthopyroxenes (b) of the Allegan chondrules.
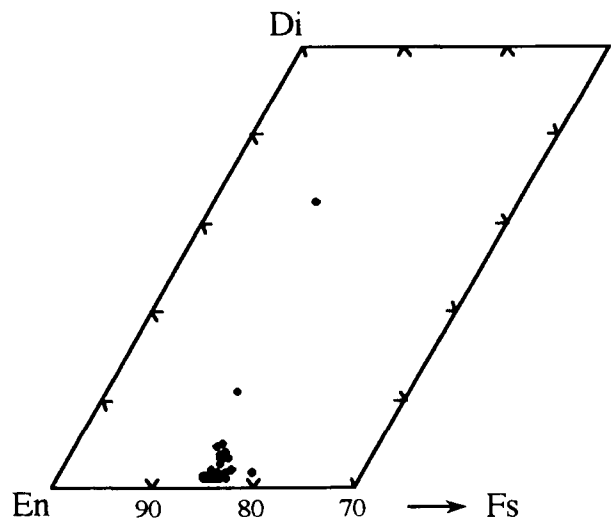

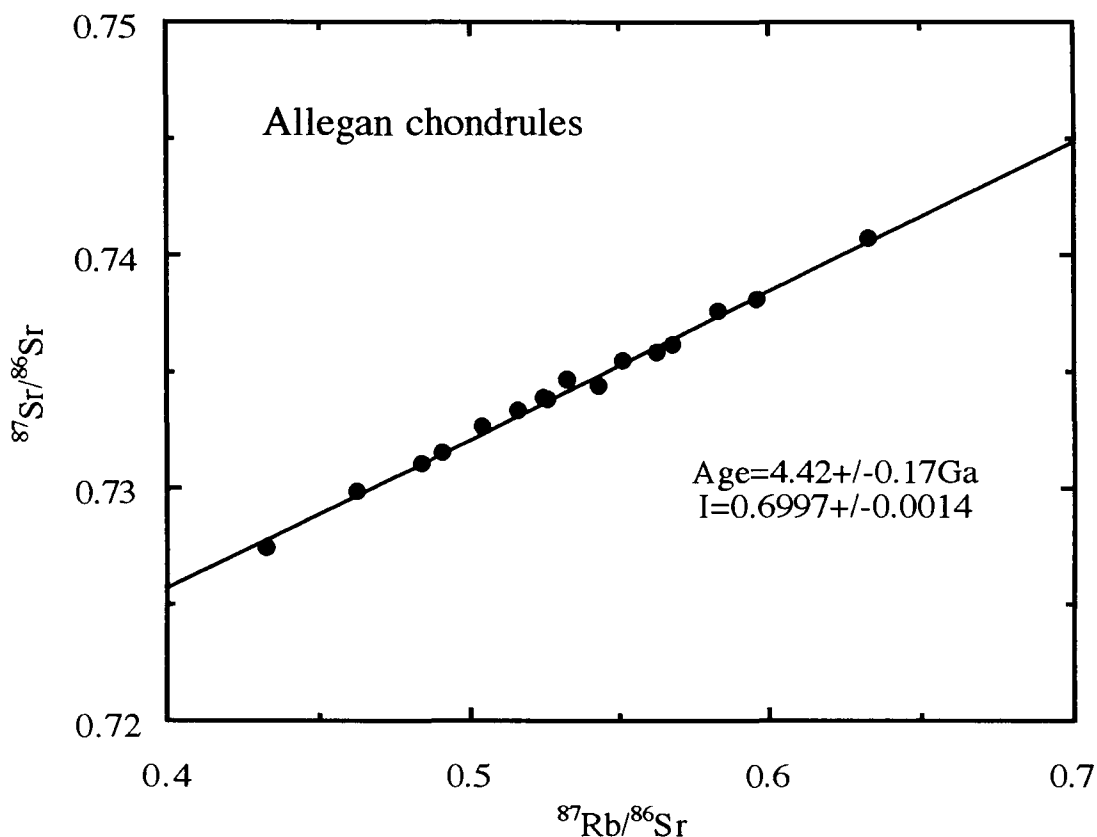Fig. 3. Pyroxene quadrilateral diagram for the Allegan chondrules.

Fig. 4. Rb-Sr isochron diagram for the Allegan chondrules. Calculation of line of best fit as per York (1969). The age was calculated using the ⁸⁷Rb decay constant of 1.42 x 10⁻¹¹ a⁻¹. Errors in age and initial ⁸⁷Sr/⁸⁶Sr ratio are indicated in 2σ.

characteristic relations between texture and Rb/Sr for the chondrules studied are consistent with the results obtained by Nishikawa (1984). An isochron age of 4.42±0.17 (2σ) Ga and an initial $^{87}Sr/^{86}Sr$ ratio of 0.6997±0.0014 (2σ) were obtained in this study (Fig. 4). The variations in Rb/Sr ratio among the chondrules, however, are remarkably small (0.149-0.218; Table 2), which could have resulted in a limited precision in the isochron age. Meanwhile, Nishikawa et al. (1990) have reported that a greenish chondrule (one of the two chondrules from the Allegan specimen they had investigated) showed a high Rb/Sr ratio (0.569). In another study on the same Allegan specimen (Nishikawa, 1984), a greenish chondrule was also found to have had the highest Rb/Sr ratio (0.594) among ten chondrules of varying lithologies. This greenish type, however, was not found in any of the chondrules from the same specimen used in this study. Nishikawa et al. (1990) further reported a Rb-Sr isochron age of 4.38±0.12 Ga with an initial ratio of 0.70004 (error not stated) for the same specimen, though the isochron was determined on only three data points (i.e. two

chondrules and one whole rock, the latter of which plots very close to one of the two chondrules in the diagram). In spite of an obvious limitation in our samples such as absence of the greenish-type chondrules, the isochron age obtained in this study is quite consistent with that of Nishikawa et al. (1990).

IV. Discussions

In the conventional sense of having constant Fe/Mg ratios in olivines and pyroxenes which are large enough for electron probe analysis (Van Schmus and Wood, 1967), the Allegan chondrite is "equilibrated", although, based on our results and the classification by Dodd (1981), it might be classified as petrologic type 4 instead of type 5. In this context, the isochron age of 4.42±0.17 Ga might be explained as the age of thermal metamorphism followed by "equilibration" as mentioned above. This interpretation also seems to well explain the general but not rigid tendency that "equilibrated" chondrites (types 4-6) show younger ages than "unequilibrated" (type 3) ones
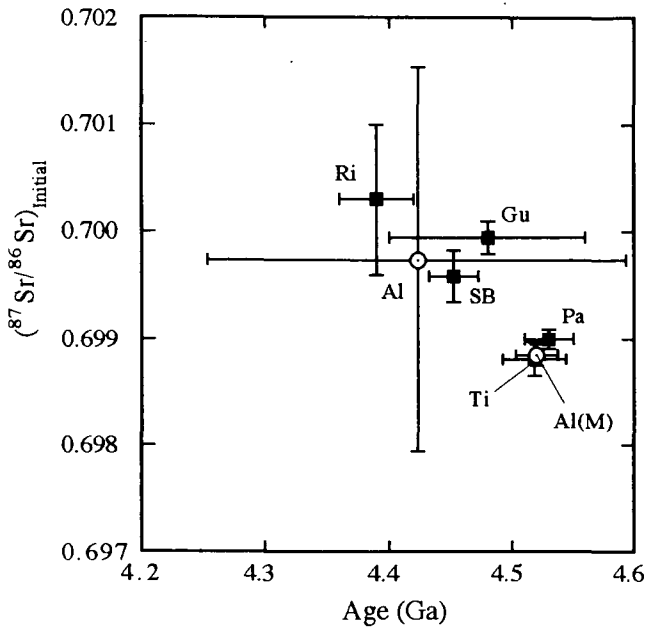
Fig. 5. Plots of Rb-Sr age vs. initial $^{87}Sr/^{86}Sr$ ratio for ordinary chondrites. Abbreviations: Ti = Tieschitz (H3) (data is taken from Minster and Allègre, 1979); Pa = Parnallee (LL3) (Hamilton et al., 1979), SB = Soko Banja (LL4) (Minster and Allègre, 1981); Ri = Richardton(H5) (Evensen et al., 1979); Gu=Guareña (H6) (Wasserburg et al., 1969); Al = Allegan (H5) (this study); Al (M) = mean model age of Allegan chondrules (this study). Ages are calculated using the decay constant of $^{87}Rb = 1.42 \times 10^{-11}$ a^{-1}.

Fig. 6. Model ages of indidual chondrules from the Allegan chondrite. The ages were calculated using an initial $^{87}Sr/^{86}Sr = 0.69885$ which was determined for ordinary chondrites by Minster et al. (1982).

(e.g. Fig. 5 in this study; Kaneoka, 1980). Some evidences from literature, however, indicate that the Allegan is an unequilibrated chondrite. Rambaldi et al. (1981) carried out analyses of ultrafine matrix ("Holy Smoke") from six ordinary chondrites including the Allegan. They found some volatile and refractory elements highly enriched in the matrix relative to the bulk chondrite. Also, some micron-sized olivine-like grains were found to contain variable Fe/Mg ratios in contrast to the constant ratio of the larger grains. Those observations clearly indicated that the Allegan chondrite as a whole had not equilibrated during and after the aggregation. This means that, if one would still regard the homogenization of Fe/Mg ratios in olivines and pyroxenes as having been due to equilibration associated with thermal metamorphism in or on the parent body/bodies, such a process (chemical equilibration among olivines and orthopyroxenes in chondrules and coarse-grained matrix) should have occurred prior to the aggregation of the materials with unequilibrated fine-grained matrix. In this case, the aggregation of materials of different origins
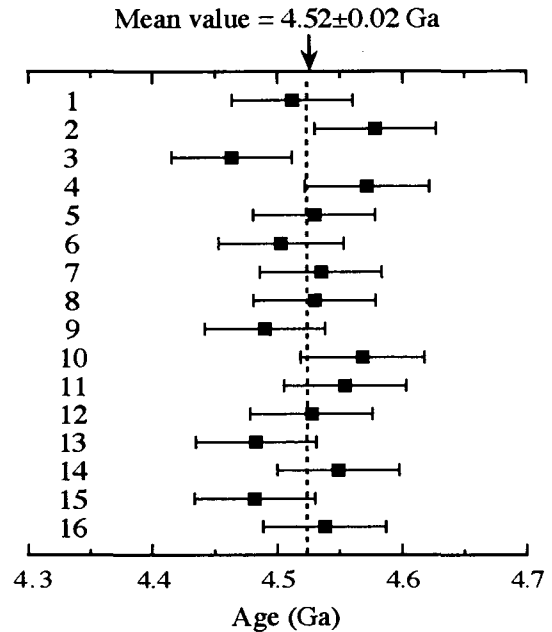
could have been caused by impact-processes on the parent body/bodies. This interpretation, however, seems unlikely because the Allegan chondrite shows minimal brecciation or shock effects (Fredriksson, 1983). Furthermore, Fredriksson (1983) found that the CaO and Al_2O_3 contents as well as the CaO/TiO_2 and Al_2O_3/TiO_2 ratios among separated chondrules from the Allegan scatter widely, while those of the "unequilibrated" Tieschitz (H3) chondrite are fairly constant. Consequently, he concluded that the Allegan chondrite is chemically unequilibrated and, therefore, should not be a metamorphic rock. Alternatively, the constant Fe/Mg ratios might have been inherited from preceding processes (e.g. chondrule formation processes). In this context, the age of 4.42 ± 0.17 Ga obtained for the Allegan chondrules in this study could imply the time of chondrule formation processes. Unfortunately, this isochron age does not have enough precision to ensure such a proposition. It can be said that analytical precision for each chondrule could be good enough but the extremely small variation in Rb/Sr ratios among all the chondrules results in such an unsatisfactory precision of the isochron age. In the meantime, the model ages for chondrules (Fig. 6) are nearly identical and thus may be more significant than the isochron age in this case. The mean value of model ages for the

sixteen chondrules, 4.53 ± 0.02 (2σ-mean) Ga, may therefore indicate more specifically the formation age of the chondrules. It is noted that as shown in Fig. 5, this model age is identical to the well-defined Rb-Sr internal isochron ages of "unequilibrated" ordinary chondrites; 4.53 ± 0.04 for the Parnallee (LL3) (Hamilton et al., 1979) and 4.52 ± 0.03 Ga for the Tieschitz (H3) (Minster and Allègre, 1979). It should be noted that the former isochron was determined from five data points for each chondrule only. The agreement in age seems to support the above interpretation that basically the Allegan chondrite might not have experienced an episode of thermal metamorphism or, even if it had, the chondrite had not been substantially affected by any such secondary processes. In this case, the chondrite might be seen as having mainly preserved the records of primary processes.

V. Conclusion

The Fe/Mg+Fe ratios of olivines and orthopyroxenes in the Allegan chondrules are considerably, but not so thoroughly, constant (Fa=17.5%, MD=1.3%; Fs=15.4%, MD= 3.2%). In the conventional sense, this meteorite thus might possibly be an H4 chondrite, even though it has already been classified as H5 based on some petrographical and chemical investigations. Furthermore, the occurrence of clinopyroxenes of endiopsidic and magnesium pigeonitic compositions suggest that this chondrite as a whole is unequilibrated chemically. As concluded from some previous investigations on the same chondrite (e.g. Fredriksson, 1983), our results suggest that this chondrite has essentially not experienced lithostatic thermal metamorphism. The homogenization of Fe-Mg compositions in the two minerals therefore could have occurred in the early processes (e.g. chondrule formation processes) before the assembly of chondrules and matrices.

The Rb-Sr internal isochron age (4.42 ± 0.17 Ga) determined from the sixteen chondrules is thus considered to be indicative of the time of such processes. The average model age for single chondrules (4.52 ± 0.02Ga) seems to define the time more precisely. Considering the good agreement of this model age with well-defined Rb-Sr internal isochron ages for "unequilibrated" ordinary chondrites, the age may imply the time of occurrence of some early processes such as chondrule formation in the solar system.

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