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MAGNETIC PROPERTIES AND NATURAL REMANENT MAGNETIZATION OF CARBONACEOUS CHONDRITES CONTAINING PYRRHOTITE

Takesi NAGATA, Minoru FUNAKI and Hideyasu KOJIMA

National Institute of Polar Research, 9–10, Kaga 1-chome, Itabashi-ku, Tokyo 173

Abstract: Magnetic properties, NRM characteristics and magnetic minerals of four carbonaceous chondrites, the Allende, the Leoville, Y-74662 and Y-81020, are examined. These C-chondrites contain ferrimagnetic pyrrhotite grains in addition to magnetite, kamacite and/or taenite as magnetic minerals possessing NRM. The low temperature NRM component which is possessed by ferrimagnetic pyrrhotite at temperatures below 300°C, indicates that the corresponding paleointensity (F_p) is around 1 Oe in order of magnitude. The high temperature NRM component possessed by magnetite and/or taenite is magnetic at temperatures below about 600°C, giving rise to $F_p \leq 0.1$ Oe. The kamacite magnetization contributes very little at temperatures below 770°C.

1. Introduction

Since carbonaceous chondrites are the most primordial undifferentiated meteorites, it is believed that their natural remanent magnetization (NRM) was acquired in the early stage of the primordial solar system formation, though some moderate effects of thermal metamorphism may have modified the original NRM to a certain extent.

One of characteristics of carbonaceous chondrites (C-chondrites) reported up to the present is that the paleointensity (F_p) of a possible ambient magnetic field during their formation process, derived from their NRM, amounts to 1 Oe or a little larger (e. g., NAGATA, 1979). Taking into consideration their experimental result that chondrules appear to consist, at least, of two components, SUGIURA *et al.* (1979) suggested that NRM of chondrules of the Allende chondrite is composed of a high temperature component which had been acquired when individual chondrules were cooled through the Curie point before they were assembled into the Allende meteorite, and the other low temperature component which was acquired in a magnetic field of about 1 Oe intensity experienced during or after the assembly of the meteorite.

Since no identification of magnetic minerals possessing the NRM was made in the previous reports, WASILEWSKI (1981) specifically pointed out a high possibility that the largest part of Allende NRM is possessed by ferrimagnetic iron-sulfide (pyrrhotite) which is thermally unstable at temperatures above 320°C, and proposed a complete revision of paleomagnetic research of Allende NRM, because he belived than Allende NRM was acquired as crystalline (or chemical) remanent magnetization (CRM) caused by a sulfidation of iron in a magnetic field. Later more extensive studies on Allende NRM, including an identification of magnetic minerals have confirmed several key points in the problem regarding to the origin of Allende NRM (NAGATA and FUNAKI, 1983). Confirmed points are as follows:

(a) Magnetic minerals are magnetite, Ni-rich (about 65 wt% Ni) taenite and pyrrhotite.

(b) NRM of both bulk and matrix specimens can be thermally demagnetized almost completely by heating to 320° C, which is close to Curie point (310° C) of ferrimagnetic pyrrhotite.

(c) The thermal demagnetization curves of unstable NRM of chondrules consist of a low temperature component which is thermally demagnetized by heating to 320° C and a high temperature one thermally demagnetized at about 600°C which is approximately equal to Curie point of magnetite and Ni-rich taenite.

(d) The TRM acquisition rate of the bulk specimen in a constant magnetic field is much smaller in the low temperature range $(25 \sim 320^{\circ} \text{C})$ than in the high temperature range $(320 \sim 600^{\circ} \text{C})$, so that the paleointensity $(F_{\rm p})$ for the low temperature component amounts to $1 \sim 3$ Oe, while $F_{\rm p}$ value for the high temperature component is smaller that 0.01 Oe.

It appears thus that a particular NRM behavior of coexistence of a low temperature component indicating $F_p = (1 \sim 3)$ Oe together with a high temperature component indicating $F_p \leq 0.01$ Oe has been confirmed for Allende C-chondrite.

In the present study, NRM characteristics of two Antarctic C-chondrites, Y-74662 (CM2) and Y-81020 (CO3), are examined in some detail on the basis of their magnetic mineralogy, together with re-examinations of Allende (CV3) and Leoville (CV3), magnetic properties of which have already been reported.

2. Magnetic Mineralogy, Magnetic Properties and NRM of Y-74662 (CM2) Chondrite

As indicated by the bulk chemical composition (HARAMURA et al., 1983), this C-chondrite contains no magnetite so that the bulk content of Fe_2O_3 is zero, but it contains a considerable amount of iron sulfide as represented by 7.38 wt% FeS in the bulk composition. Results of EPMA analyses of six iron sulfide grains in a thin section of this chondrite are given by $S/Fe=1.092\pm0.028$ in atomic ratio. In addition to the iron sulfide, this chondrite contains kamacite grains of Ni/(Fe+Ni)= $0.044\pm$ 0.002 in atomic ratio. Figure 1 shows an example of the first run thermomagnetic cycle curve which was measured under the steady magnetic field at 10 kOe in the 10^{-4} torr atmospheric pressure, where a low apparent Curie point takes place at 295°C and a high apparent Curie point at 620°C. A small residual magnetization having Curie point at 770°C is also detectable. The low Curie point disappears almost completely in the cooling curve, suggesting that the magnetic phase corresponding to the low Curie point is destructed by the first run heating to 800°C. Since Curie point (Θ_c) and the upper stability temperature limit (T') of ferrimagnetic monoclinic pyrrhotite are $\Theta_c = 310^{\circ}$ C and $T' = 254^{\circ}$ C respectively (e. g., CRAIG and SCOTT, 1976), it seems most likely that the magnetic phase corresponding to the low apparent Curie point is ferrimagnetic pyrrhotite. The high apparent Curie point at 620°C would be considered to correspond to either taenite or magnetite, but neither of the two



Fig. 1. The first-run thermomagnetic curve of Y-74662 (CM2) (external magnetic field=10 kOe).

Item	Y-74662 (CM2)	Y-81020 (CO3)	Allende (CV3)	Leoville (CV3)
$I_{\rm s}$ (emu/g)	1.08	14.0	1.31	10.3
$I_{\rm R}$ (emu/g)	0.103	1.39	0.26	0.58
$H_{\rm C}$ (Oe)	120	170	175	35
$H_{\rm CR}$ (Oe)	735	500	530	225

Table 1. Magnetic hysteresic cycle paramenters.

minerals was detected by the present EPMA survey over isolated mineral grains larger than 10 μ m in diameter. All detected Fe-Ni metallic grains kamacite.

It appears likely therefore that taenite fine grains, considerably smaller than 10 μ m in diameter, may be present in matrix of this chondrite. The composition of the taenite would be about 50 Ni at% corresponding to 620°C of Curie point.

In Table 1, magnetic hysteresis cycle parameters, saturation magnetization (I_s) , saturated isothermal remanent magnetization (I_R) , magnetic coercive force (H_C) and remanence coercive force (H_{RC}) at room temperature $(20 \sim 24^{\circ}C)$ of a bulk specimen of Y-74662 chondrite are listed. Considerably large values of $H_{RC}=735$ Oe and $I_R/I_s=0.095$ may suggest that this chondrite contains magnetically coercive grains having $H_C \sim 5 \times 10^2$ Oe (NAGATA and CARLETON, 1987).

Figure 2 illustrates a set of (I) thermal demagnetization curve of NRM and (II) a partial thermoremanent magnetization (PTRM) acquisition curve in a magnetic field of 0.50 Oe of a bulk specimen of Y-74662 chondrite. As shown in the figure, about 85% of NRM is thermally demagnetized at about 300°C, and practically no residual NRM remains at temperatures above 600°C, while PTRM acquisition rate markedly increases at temperatures above 300°C. This Y-74662 NRM can be divided



Fig. 2. (1) (Left) Thermal demagnetization curve of NRM. (II) (Right) PTRM acquisition curve. Y-74662 (CM2).



Fig. 3. Königisberger diagrams of Y-74662 (CM2). (Left) Low temperature component. (Right) High temperature component.

at about 300°C into a low-temperature component and a high temperature one. The directions of the high temperature component and the low temperature one are in approximate agreement with each other within $\pm 20^{\circ}$ in deviation angle.

Figure 3 shows the Königisberger diagrams to estimate paleointensity for NRM (F_p) on the basis of Königisberger-Thellier experiment, separately for the low and high temperature components. The estimated F_p values are 0.82 Oe for the low temperature component and 0.011 Oe for the high temperature component.

It seems very likely that NRM of Y-74662 (CM2) chondrite also is composed of a low temperature component which is attributable to TRM acquired by ferrimagnetic pyrrhotite grains in a magnetic field of $F_p \sim 1$ Oe, and a high temperature component attributable to TRM probably acquired by taenite grains in a magnetic field of $H_p \sim$ 0.01 Oe, just as in the case of Allende (CV3) chondrite (NAGATA and FUNAKI, 1983).

3. Magnetic Mineralogy, Magnetic Properties and NRM of Y-81020 (CO3) Chondrite

NRM of another Antarctic C-chondrite, Y-81020 (CO3), also consists of more than two components. Magnetic hysteresis cycle parameters and the first run thermomagnetic cycle curves are shown in Table 1 and Fig. 4 respectively.

As briefly described in the photographic catalog of the Antarctic meteorites (YANAI and KOJIMA, 1987), this chondrite contains magnetic opaque minerals such as iron sulfide and Fe-Ni metals in its matrix. EPMA analyses of 22 iron sulfide grains larger than 10 μ m in diameter are summarized as a mean atomic ratio of S to Fe given by S/Fe=1.025±0.015. Detected Fe-Ni metallic grains are only kamacite, composition of which is given by Ni/(Fe+Ni)=0.053±0.005 in atomic ratio. No taenite of grain size larger than 10 μ m is detected.

The first run thermomagnetic cycle curves illustrated in Fig. 4 show Curie point of taenite and/or magnetite at 580°C, and $\alpha \rightarrow \gamma$ transformation point of kamacite at 770°C in the heating process, and $\gamma \rightarrow \alpha$ transformation at 600°C in the cooling process, but no definite evidence of presence of ferrimagnetic pyrrhotite is detectable. However, the thermal demagnetization curve of NRM of this chondrite shown in Fig. 5 suggests that NRM of this chondrite is composed of a low temperature component having the upper limit at about 350°C, an intermediate component extending below 580°C, and a high temperature component extending above 580°C. A plausible interpretation of the NRM structure would be such that the low, intermediate and high components represent respectively ferrimagnetic pyrrhotite, taenite and/or magnetite and kamacite.

Figure 6 shows a Königisberger diagram to estimate paleointensity F_p for this chondrite. Fluctuating deviations of individual plots from an assumed linear relation between the residual NRM and the PTRM acquisition may not be negligibly small



Fig. 4. The first-run thermomagnetic curve of Y-81020 (CO3) (external magnetic field=10 k Oe).

in this case. Roughly speaking, however, and approximate linear relation between the residual NRM and the PTRM acquisition for the low temperature range (below 300° C) can lead to an approximate paleointensity value as $F_{\rm p} \sim 1.2$ Oe. In the same way, an approximate paleointensity of the intermediate temperature component (for the range between 300° C and 580° C) is $F_{\rm p} \sim 0.09$ Oe. The paleointensity of the



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high temperature component is numerically evaluated as $F_{\rm p} \sim 0.006$ Oe.

In the case of Y-81020 chondrite also, the direction of the low temperature NRM component and that of the intermediate temperature component are in approximate agreement with each other, with fluctuating deviations smaller than $\pm 30^{\circ}$ in angle from the median values.

4. Magnetic Mineralogy, Magnetic Properties and NRM of Allende (CV3) and Leoville (CV3) Chondrites

Magnetic properties and NRM characteristics of Allende (CV3) chondrite have already been reported (NAGATA and FUNAKI, 1983), and those of Leoville (CV3) chondrite also have been outlined (NAGATA and SUGIURA, 1976, 1977). Only key points of these previous works will be summarized here, together with some new additional experimental results.

Typical examples of measured magnetic hysteresis cycle parameters of these two chondrites are listed in Table 1. It is noted that ratio $I_{\rm R}/I_{\rm s}$ of the Allende amounts to $I_{\rm R}/I_{\rm s}=0.20$, which represents a magnetic constitution of this chondrite containing a considerable share of a high coercive component, such as ferrimagnetic pyrrhotite.

The first run thermomagnetic curves of both Allende and Leoville chondrites show no definite magnetic transition point around 300°C. The thermomagnetic curve of the Allende represents almost completely a single magnetic phase of magnetite and/or taenite having Curie point around 610°C, and that of the Leoville looks likely to consist of a magnetic phase of magnetite and taenite of around 575°C in Curie point (85%) and a kamacite of about 760°C in its $\alpha \rightarrow \gamma$ transition temperature (15%).

However, the thermal demagnetization curves of NRM, obtained in 10^{-5} torr atmospheric pressure, of both the Allende and the Leoville clearly indicate that more than 50% of their NRMs are thermally demagnetized at 300°C, as shown in Figs. 7 and 8 respectively. NRM of the Allende is almost completely thermally demagnetized by heating to 300°C, as in the case of Y-74662, while about 30% of the Leoville NRM still remains after thermally demagnetizing up to 300°C, in a similar way to the case of Y-81020.

In Table 1, on the other hand, I_s values are 1.08 emu/g and 1.31 emu/g in Y-74662 and the Allende respectively, while they are much larger in Y-81020 and the Leoville, amounting to 14.0 emu/g and 10.3 emu/g respectively. The observed larger value of relative occupation by the high temperature NRM component in the second group (*i. e.*, Y-81020 and the Leoville) may correspond to most parts of the observed larger value of I_s of this group.

The paleomagnetic field intensity (F_p) obtained with the aid of königisberger-Thellier technique from the Allende ranges between 1 and 3 Oe for the low temperature component (20~300°C) and about 0.01 Oe in order of magnitude for the high temperature component (300~560°C). The paleomagnetic field intensity of the low temperature NRM component of the Leoville is evaluated as $F_p=0.97$ Oe, and that of the high temperature component (300~560°C) is given by $F_p=0.05$ Oe.

The Allende contains iron-sulfide, taenite and magnetite grains, no kamacite grain being detected by EPMA analyses. The compositions of iron-sulfide and taenite



analyzed by EPMA technique are given by $S/Fe=1.089\pm0.006$ in atomic ratio for the mean value of 4 iron-sulfide grains and $Ni/(Fe+Ni)=0.684\pm0.010$ in atomic ratio for the average value of four taenite grains. It will be certain in the Allende that the low temperature NRM component is possessed by ferrimagnetic pyrrhotite grains and the high temperature one by Ni-rich taenite and magnetite grains.

A thin section sample of the Leoville for the purpose of an EPMA analysis has

not yet been available in the writers' laboratory. Judging from the characteristics of the thermomagnetic curve (NAGATA and SUGIURA, 1976) and the thermal demagnetization curve of NRM (Fig. 8) of this chondrite, it seems likely that the low temperature NRM component is possessed by ferrimagnetic pyrrhotite, and the high temperature one by taenite and magnetite.

5. Iron Sulfide in Carbonaceous Chondrites

As described in the preceding sections, the low temperature NRM component of all the four C-chondrites, Y-74662, Y-81020, the Allende, and the Leoville, is possessed by a ferro- or ferrimagnetic mineral grains, magnetic transition temperature of which is about 300°C. Among ferromagnetic or ferrimagnetic minerals which are so far known to be present in meteorites, only ferrimagnetic pyrrhotite and ferromagnetic tetrataenite have their magnetic transition temperature at about 300°C, below which they can acquire remanent magnetization by an appropriate magnetization process. Only a known C-chondrite which contains tetrataenite is Y-791717 (CO3) chondrite (NAGATA and CARLETON, 1989). Because of its extremely large magnetic coercivity and its practically irreversible transformation to disordered taenite at temperatures higher than 310°C, the tetrataenite phase is clearly detected with the



FeS in C - chondrites

Fig. 9. Histograms of S/Fe atomic ratio of iron-sulfide grains in four C-chondrites.

aid of magnetic analysis technique. It seems very likely therefore that the low temperature NRM components in four C-chondrites in the present study are possessed by ferrimagnetic pyrrhotite grains. For the purpose of examining the composition of iron sulfide minerals in Y-74662, Y-81020 and the Allende EPMA analyses of their thin sections were carried out in some detail.

Figure 9 shows histograms of S/Fe in atomic ratio of iron sulfide grains in the Allende, Y-74662 and Y-81020 in comparison with a histogram of S/Fe of iron sulfide grains in Y-791717 which contains only troilite. The median values of S/Fe histograms of both the Allende and Y-74662 are close to the stoichiometric composition of ferrimagnetic monoclinic pyrrhotite, Fe_7S_3 . The S/Fe histogram of Y-81020 also is shifted to the ferrimagnetic structure composition.

Since the crystal structure of iron sulfide is subjected to a complicated equilibrium phase system associated with various magnetic properties within a composition range of S/Fe between S/Fe=1.0 and S/Fe=1.15 (e.g., CRAIG and SCOTT, 1976), it may be difficult to determine exact magnetic properties of the examined iron sulfide grains in the three C-chondrites from their chemical structures shown in Fig. 9 only.

It may be possible to conclude, however, that all results of mineralogical and magnetic analyses of the three C-chondrites are in favor of ferrimagnetic iron sulfide grains (ferrimagnetic pyrrhotite) as the carrier of their low temperature NRM component.

6. Stability and Reproducibility of NRM of C-Chondrites

In Table 2, the main and auxiliary magnetic minerals, NRM intensity and its allotment over its blocking temperature (T) range, and the paleomagnetic field intensity (F_p) derived separately from the low and high temperature components of NRM of the four C-chondrites are summarized. The classification of magnetic minerals in the table is based on the total results of wet chemical analysis of bulk composition, EPMA analysis of individual opaque minerals, and magnetic analysis of both thermomagnetic curves and NRM thermal demagnetization curve. Hence

Item	Y-74662 (CM2)	Y-81020 (CO3)	Allende (CV3)	Leoville (CV3)
Main mag. mineral Auxiliary mag. mineral	FeS _{1+χ} Fe-Niγ Fe-Niα	Fe₃O₄ FeS₁+χ Fe-Niα	Fe ₃ O ₄ FeS _{1+X} Fe-Niř	Fe_3O_4 $FeS_{1+\chi}$
NRM intensity	5.9×10-4	8.6×10 ⁻⁴	$(0.8 \sim 2.5) \times 10^{-4}$	1.2×10^{-4}
Low temp. comp.	$T \leq 300^{\circ} \text{C}$	<i>T</i> ≤320°C	$T \lesssim 320^{\circ} \mathrm{C}$	<i>T</i> ≲300°C
High temp. comp.	(86%) 300 <i><t<< i="">700°C (13%)</t<<></i>	(60%) 320< <i>T</i> <640°C (36%)	(≥95%) 320< <i>T</i> <700°C (<5%)	(66%) 300 <i><t<< i="">560°C (19%)</t<<></i>
Low temp. F_p (Oe)	0.82	1.2	0.8 3.0	0.97
High temp. F_p (Oe)	0.011	0.09	<0.06	0.05

Table 2. Magnetic mineralogy and NRM characteristics.





the classification is not exactly quantitative.

The paleointensity (F_{p}) is determined on an assumption that NRM was acquired with the thermoremanent magnetization (TRM) acquisition process in a magnetic field, F_{p} . The TRM acquisition assumption can be considered to be a highly plausible process, even though it may not be the unique solution for possible NRM acquisition mechanism. In the case of the Allende, for example, Fig. 7 illustrates the thermal demagnetization curves of partial TRM (PTRM) acquired by cooling from 320°C to 20°C in a magnetic field of 0.44 Oe, together with the thermal demagnetization curves of NRM; the decreasing rate of the remanent magnetization with an increase in temperature is approximately proportional between PTRM and NRM. On the other hand, Fig. 10 shows and approximately linear relationship between the PTRM intensity acquired by cooling from 320°C and an applied magnetic field for three bulk specimens of the Allende. These two experimental results may support the plausibility of TRM acquisition assumption for the origin of the low temperature NRM component of the Allende. Strictly speaking, the assumed positive linear relationship between the thermal demagnetization curve of NRM and that of PTRM or the assumed negative linear relationship between the thermal demagnetization curve of NRM and the PTRM acquisition curve in a constant weak magnetic field for carbonaceous chondrites is accompanied by fluctuations of a considerable magnitude, as shown in Fig. 3 and Fig. 6 for example.

Figure 11 shows a comparison of the thermal demagnetization rates of NRM within successive temperature intervals, $\Delta T = T_i + 1 - T_i$, with the PTRM acquisition rates within the same temperature intervals, ΔT , for the low temperature NRM range of the three carbonaceous chondrites, where the PTRM intensity is normalized to the case of applied magnetic field=1 Oe. As shown in the figure, the total intensity of the low temperature NRM component is approximately equal to the total TRM acquired during cooling from 300°C to 0°C in a magnetic field of 1 Oe intensity for



BLOCKING TEMPERATURE SPECTRUM

Fig. 11. Spectrum of NRM thermal demagnetization rate with respect to temperature (left side), and spectrum of PTRM acquisition rate with respect to temperature in the normalized magnetic field of 1.0 Oe (right side), for Allende (CV3), Y-74662 (CM2) and Y-81020 (CO3).

the three examined C-chondrites, as far as order of magnitude of remanent magnetization is concerned. As for the expected individual proportional correspondence between the thermal demagnetization rate of NRM within individual ΔT and the PTRM acquisition rate within ΔT , however, considerably large fluctuations are involved in all the three examples. The large fluctuations are considered to be due to an instability of the structure of iron sulfide crystals themselves, in part, and also in part to poor reproducibility of ferrimagnetic iron sulfide phases owing to the complicated phase equilibrium conditions among various phases such as Fe_7S_8 (Curie point $\simeq 300^{\circ}C$), $Fe_{\theta}S_{10}$, etc. (Curie point=270~210°C) (e.g., O'REILLY, 1984). Experimental tests to estimate the reproducibility of thermal demagnetization of NRM and acquisition of PTRM for the low temperature range (i. e. $T \leq 310^{\circ}$ C) were carried out with respect to the Allende and Y-81020. In both C-chondrites, the mean values of fluctuating deviations of observed values from the average thermal demagnetization curve of NRM and from the average PTRM acquisition curves, respectively, are within 30% (e.g., NAGATA and FUNAKI, 1989). The mechanism for the stability of remanent magnetization with respect to temperature changes and the reproducibility of PTRM acquisition in the iron-sulfide grains in C-chondrites has not yet been completely clarified. A comparison of the spectrum of NRM thermal demagnetization with respect to increasing temperature with that of PTRM acquisition with respect to decreasing temperature, illustrated in Fig. 11, would be one of the best possible ways to express mutual differences as well as similarity between the NRM and the PTRM of C-chondrites.

7. Concluding Remarks

In the present study, it is experimentally confirmed that not only the Allende (CV3) chondrite but Y-74662 (CM2) and Y-81020 (CO3) chondrites also have NRM component possessed by ferrimagnetic pyrrhotite grains having the upper limit Curie point near 300°C. The Leoville (CV3) chondrite also looks very likely to have nearly the same NRM characteristics as Y-81020.

The NRM of these four C-chondrites consists of the high temperature component which is ferromagnetic at temperatures higher than 300°C and the low temperature one possessed by ferrimagnetic pyrrhotite. The high temperature NRM component is possessed by magnetite and/or disordered taenite, Curie points of which are around 600°C. In addition, Y-81020 and Y-74662 contain a small amount of kamacite NRM.

The paleointensity (F_p) derived from the low temperature NRM component is around 1 Oe in order of magnitude, while the F_p value derived from the high temperature component is smaller than 0.1 Oe in all the four C-chondrites, provided that the origin of these NRMs assumes the TRM acquisition mechanism.

Although the conformality between the distribution spectrum with respect to temperature for the thermal demagnetization curve of the low temperature NRM component and that for PTRM produced in the same temperature range appears considerably disturbed by fluctuations, probably because of insuffciently equilibrated condition among various different ferrimagnetic phases in iron sulfide grains, the estimated order of magnitude, $F_p \sim 1$ Oe, for the low temperature component, does not seem to be very much different from the true paleomagnetic field intensity. A problem here will be that no other chondrite, except the C-chondrites, indicating $F_p \sim 1$ Oe has been found so far. Another problem is concerned with a possibility of a special physical mechanism to make ferrimagnetic pyrrhotite grains to acquire NRM, the intensity of which is evaluated by the Königisberger-Thellier technique to be much larger than the real ambient magnetic field during the formation of the examined NRM.

The problems of the NRM characteristics of the low temperature component and their relation to those of the high temperature component in the iron-sulfide containing C-chondrites may still need further studies in detail.

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