

Mineralogy and noble-gas signatures of the carbonate-rich lithology of the Tagish Lake carbonaceous chondrite: evidence for an accretionary breccia

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Abstract

The carbonate-rich lithology of the Tagish Lake carbonaceous chondrite was characterized by noble-gas mass spectrometry, synchrotron X-ray diffraction analysis, and transmission and scanning electron microscopy. Noble-gas analysis was performed on two samples and the results showed that primordial noble gases are abundant and solar noble gases are absent in the samples of carbonate-rich lithology. The concentrations of Ne-A2 and -E in both samples are at the maximum level observed for CI and CM chondrites, suggesting high abundances of presolar diamonds and SiC/graphite, respectively. The cosmic-ray exposure age cannot be determined precisely, because the shielding depth of our Tagish Lake samples is unknown, but the minimum exposure age was determined to be 5.5 ± 0.7 Myr on the basis of cosmogenic ^{21}Ne concentrations and the highest ^{21}Ne production rate. X-ray and electron-microscopic study showed that the carbonate-rich lithology is dominated by loosely packed porous matrix that consists mainly of fine-grained saponite and ferromagenesian carbonate. The matrix contains very few chondrules, but many fine-grained clasts having angular shape with longest dimensions up to 1 mm. The clasts differ from host matrix in both texture and mineralogy. They are massive, compacted material with porosity much lower than matrix and contain abundant magnetite and a coherent intergrowth of serpentine and saponite that is rare in matrix. The presence of texturally and mineralogically distinct clasts indicates that the carbonate-rich lithology is a breccia, but the absence of solar noble gases and impact-induced deformational features in host matrix distinguish it from an asteroid regolith breccia. Our results instead indicate that it is an accretionary breccia formed by simultaneous accretion of diverse objects in a massive dust cloud. The clasts often enclose chondrules and anhydrous silicate fragments such as low-iron-manganese-enriched olivines. This observation and their highly compacted, angular morphology suggests that the clasts derive from earlier-formed chondritic materials which have experienced compression and fragmentation due to mild impacts and from which they were excavated and incorporated into the final rock. It is thus suggested that, in the course of planetesimal formation, agglomeration and disagglomeration of

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small parent bodies occurred repeatedly in a dense dust cloud where solar wind from the infant sun was shielded. The fine-grained ferromagnesian carbonates must have precipitated from aqueous solutions after the assembly of the Tagish Lake asteroid from recycled materials from previous bodies, because the carbonates coat the surfaces of pores and fill in veins in the clasts.

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1. Introduction

Tagish Lake is a newly fallen (January, 2000) carbonaceous chondrite with carbon and major element concentrations close to CI [1]. It contains chondrules and CAIs that show evidence of extensive aqueous alteration and consists predominantly of two mineralogically different lithologies, carbonate-rich and -poor [2]. Transmission electron microscopy (TEM) showed that the major constituent minerals in the matrix are the hydrous silicates saponite and serpentine [2]. Oxygen isotopic data are distinct from any group of carbonaceous chondrites, but are closer to CI than to CM [3]. These mineralogical and isotopic signatures show that Tagish Lake is a new type-2 carbonaceous chondrite [2]. The meteorite exhibits a reflectance spectrum similar to that observed from the D-type asteroids [4], which are principally located in the outer asteroid belt and whose meteorite counterparts have not previously been recognized. Therefore, Tagish Lake is probably the first meteorite that preserves a record of the formation and evolution of material in the outer solar system.

We have characterized mineralogical and noble-gas isotopic signatures of the carbonate-rich lithology of the Tagish Lake meteorite. The mineralogy of fine-grained components was investigated by X-ray diffraction using synchrotron radiation and TEM. It will be shown in this paper that the unique mineralogy and texture of the carbonate-rich lithology was produced via accretion processes in the nebula and aqueous alteration reactions in an asteroid.

2. Experimental procedures

A fresh small chip (18.9 mg) was chosen for

noble-gas analysis and a large chip (~ 1 g) for mineralogical and noble-gas characterization. Five 100- μm sized matrix particles were separated from the small chip and analyzed by X-ray diffraction. The results showed that all particles are rich in carbonate, indicating that the small chip is dominated by the carbonate-rich lithology. We analyzed noble gases in the small chip without any pre-analysis processing such as rinsing in water or ethanol, to minimize terrestrial alteration – this meteorite is very friable and unstable in liquids. The large chip consists entirely of the carbonate-rich lithology, which was confirmed by the observation of three slices ($5 \times 5 \times 1$ mm in size for each slice), cut out from the chip, using a scanning electron microscope (SEM JEOL JSM-5800LV). Ne and Ar isotopic ratios and concentrations of a part (3.5 mg) of one slice were analyzed using a noble-gas mass spectrometer. Mineralogical characterization was carried out on the remaining two slices. Modal analysis of chondrules, clasts, and matrix was performed on a whole view of the two slices using the LIA32 software that measures areal ratios between objects in images. Major element concentrations in minerals and mineral aggregates were determined using an electron microprobe (EPMA JEOL JXA-733 superprobe) equipped with a wavelength-dispersive X-ray spectrometer (WDS). The WDS quantitative analyses were performed at 15 kV accelerating voltage and 10 nA beam current with a focused beam 2 μm in diameter or a defocused beam 10 μm in diameter, as required. Detailed EPMA analysis procedures are described elsewhere [5].

Based on the textural and compositional characterization by electron microscopy, 24 small lumps approximately 100 μm in size (fourteen, seven, and three from matrix, clasts, and chon-

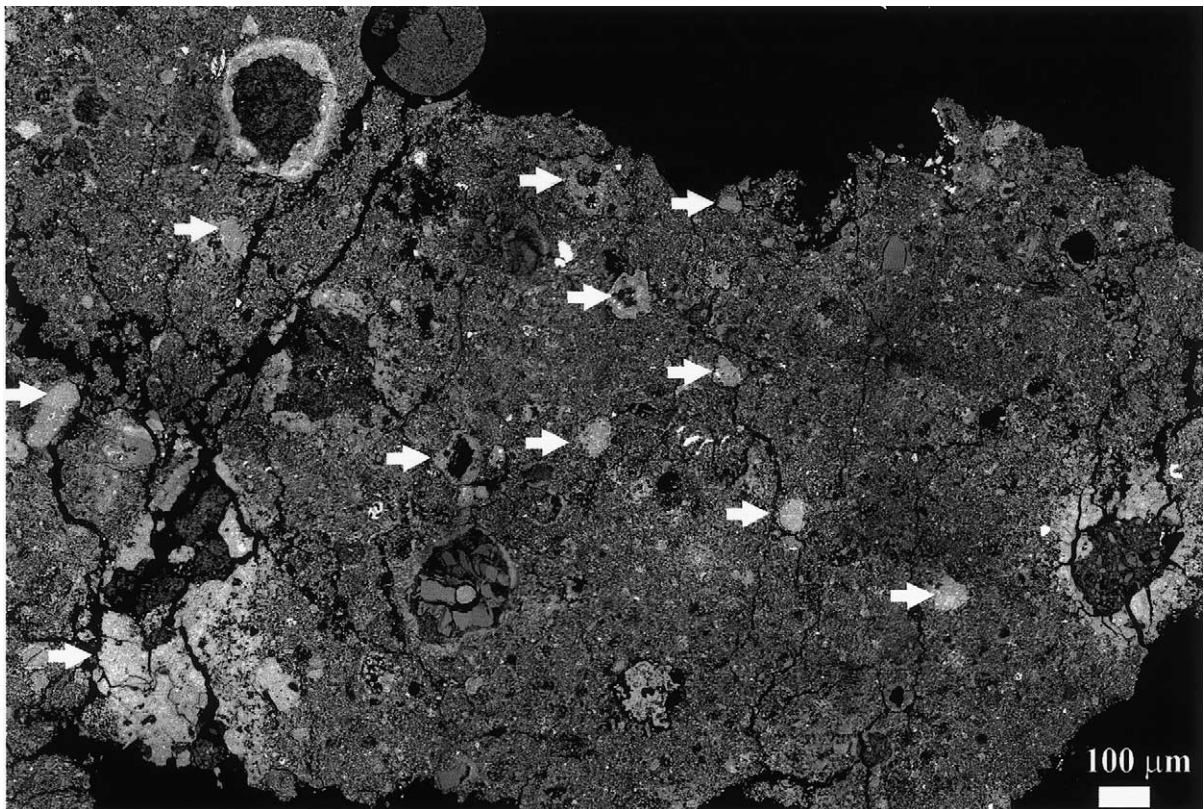


Fig. 1. A whole view of the carbonate-rich lithology of Tagish Lake meteorite. Fine-grained matrix is dominant. Chondrules and related objects occur in places in the matrix. Fine-grained angular clasts occur in many places in matrix. Some of the clasts are indicated by arrows.

drules, respectively) were separated from the portions of interest in the meteorite slices for X-ray diffraction and TEM analysis. For the X-ray diffraction analysis, individual small lumps were mounted on a thin glass fiber 5 μm in diameter and exposed to synchrotron X-rays in a Gandolfi camera. The X-rays were monochromated to $2.161 \pm 0.001 \text{ \AA}$. The Gandolfi method [6] is very useful for characterization of small minerals in meteorites (e.g., [5,7]). In the present study we applied this method to synchrotron radiation at beam line 3A of the Photon Factory Institute of Material Science, High Energy Accelerator Research Organization (Tsukuba, Japan). The ultrahigh intensity and well-monochromated synchrotron X-rays allows us to obtain a clear X-ray powder diffraction pattern of each small lump with a short exposure

duration of approximately 30 min (as compared to 24–48 h with a standard X-ray generator). After the X-ray diffraction measurements, the small lumps were embedded in epoxy resin and microtomed by a Leitz-Reichert Super Nova ultramicrotome for TEM (JEOL JEM-2000FX II) observation and quantitative chemical analysis by EDS (Philips DX4) equipped in the TEM (see [8] for detailed TEM procedures).

Noble-gas analyses were made on two samples, a chip (18.9 mg) and a slice (3.5 mg), using a MM5400 mass spectrometer at Kyushu University equipped with a Ta furnace and a stainless steel purification line called ‘Jack and the beanstalk’. Noble gases in the chip of Tagish Lake were characterized by pyrolysis at seven temperature steps with 15-min heating duration; 260,

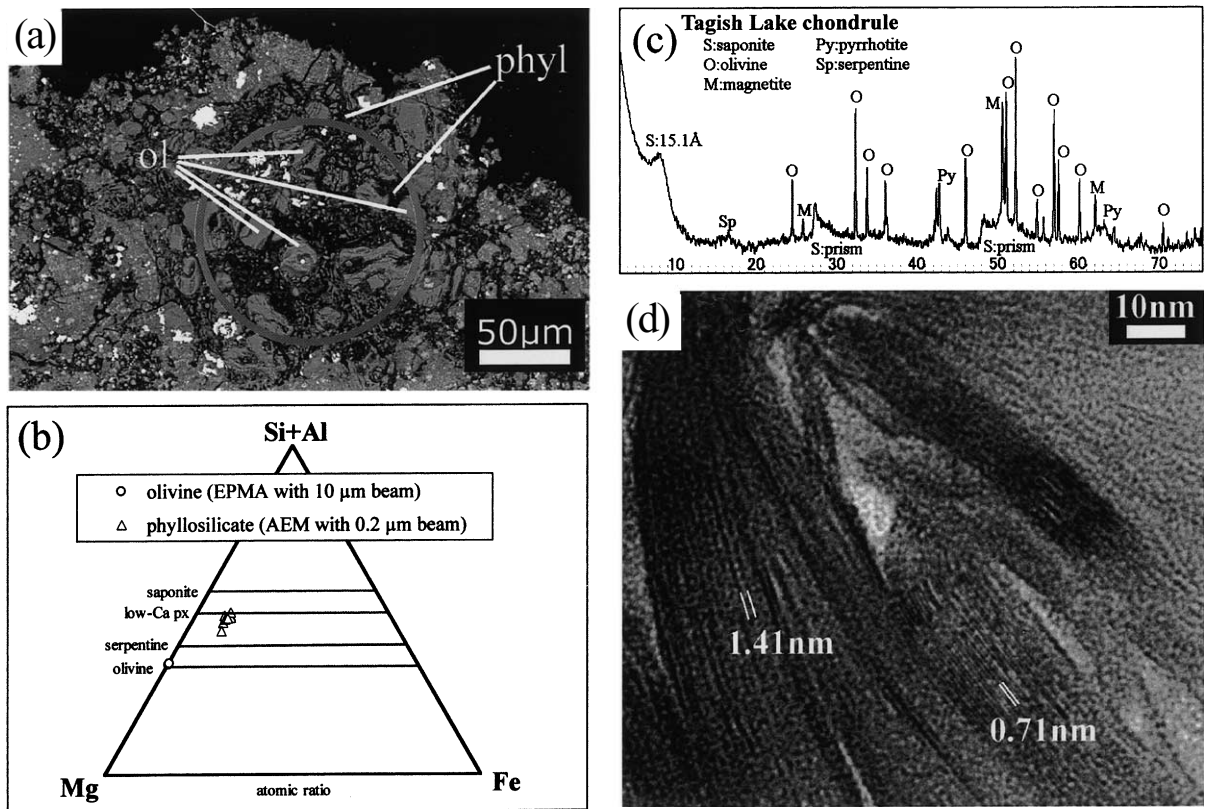


Fig. 2. Results of detailed characterization of a chondrule in the carbonate-rich lithology. (a) A BSE image showing that a part of the chondrule. Olivine and phyllosilicates are major constituents. The circle shown is an approximate portion that was later analyzed by X-ray diffraction and TEM. (b) Chemical composition of olivine and phyllosilicates in the chondrule determined by EPMA and AEM analyses, respectively. Phyllosilicate compositions are plotted between serpentine and saponite lines, suggesting that it is a mixture of these two minerals. (d) An X-ray diffraction pattern of a portion encircled in (a), showing the chondrule consists of saponite, olivine, pyrrhotite, and minor amounts of serpentine and magnetite. (d) A TEM image of phyllosilicates, showing that they are intimate mixtures of saponite (1.41 nm interlayer spacing) and serpentine (0.71 nm).

500, 680, 910, 1140, 1400, and 1700°C, while those in the slice were analyzed at 800, 1100, and 1700°C.

3. Texture and mineralogy

The carbonate-rich lithology (Fig. 1) consists of sparse chondrules and many angular to sub-rounded clasts set in a fine-grained carbonate-rich matrix. Modal analysis of a cross section of two meteorite slices showed that matrix constitutes 83 areal %, and the remaining 17 areal % is chondrules and clasts. Thus, matrix is the pre-

dominant component in this lithology, similar to most CM2 and CI1 chondrites.

3.1. Chondrules

Chondrules are rare, relatively small, ranging in size from 80 to 500 μm, and consist mainly of anhydrous and hydrous silicates. The areal ratio of anhydrous to hydrous silicates varies between chondrules. We have made detailed characterization of several chondrules and the results of a typical one are shown in Fig. 2a–d. This chondrule is composed mainly of olivine phenocrysts and phyllosilicates that fill spaces between olivines

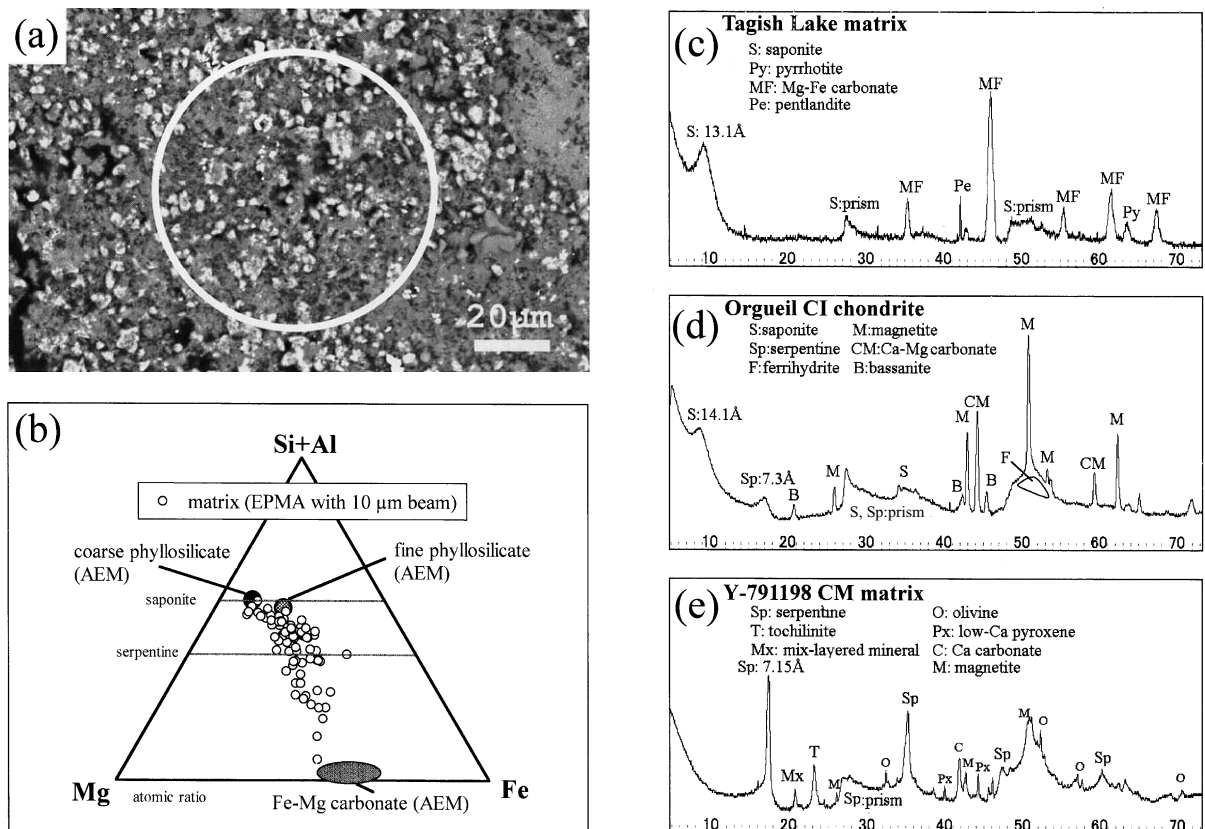


Fig. 3. Texture, mineralogy, and chemical composition of matrix in the carbonate-rich lithology. (a) A BSE image of a portion of matrix, showing that it is a porous aggregate of fine Mg-Fe carbonate (white grains in the figure) and saponite (gray). The circle shows the portion investigated by X-ray diffraction and TEM. (b) Chemical composition determined by EPMA and AEM analyses. Since the 10- μm beam is much larger than individual minerals in matrix (see (a)), each EPMA analysis in the figure indicate average compositions of a 10- μm area, while AEM data indicate compositions of nearly pure mineral phases present in the matrix because 0.3- μm beam was used for AEM analysis. The average matrix composition is near the center of the diagram (not plotted), suggesting that the abundance ratio of saponite and Mg-Fe carbonate in matrix is approximately 3:2. (c) An X-ray diffraction pattern of matrix of carbonate-rich lithology of the portion encircled in (a), showing saponite and Mg-Fe carbonate are major phases. (d) An X-ray diffraction pattern of Orgueil CI chondrite. Major phases are saponite, serpentine, magnetite, and Ca-Mg carbonate. (e) An X-ray diffraction pattern of matrix of Y-791198 CM2 chondrite. Major phases are serpentinite and tochilinite. The three (c–e) X-ray diffraction patterns are quite different.

(Fig. 2a). The olivines are almost pure forsterite and the phyllosilicates have compositions intermediate between saponite and serpentine (Fig. 2b). Synchrotron X-ray diffraction of the portion encircled in Fig. 2a showed that it consists mainly of olivine and saponite with minor amounts of serpentine, pyrrhotite, and magnetite (Fig. 2c). TEM observation of the portion analyzed by X-ray diffraction showed that phyllosilicates of the chondrule are intimate mixtures of saponite and serpentine (Fig. 2d).

3.2. Matrix

Matrix is very porous aggregate of fine mineral particles (Fig. 3a). Analytical electron microscope (AEM) analysis using a 0.3- μm beam identified three mineral phases: fine- and coarse-grained phyllosilicates and ferromagnesian (Mg-Fe) carbonates (Fig. 3b). EPMA analysis using a 10- μm beam showed that matrix compositions fall within regions defined by the mineral phases identified by AEM, indicating that matrix consists mainly of

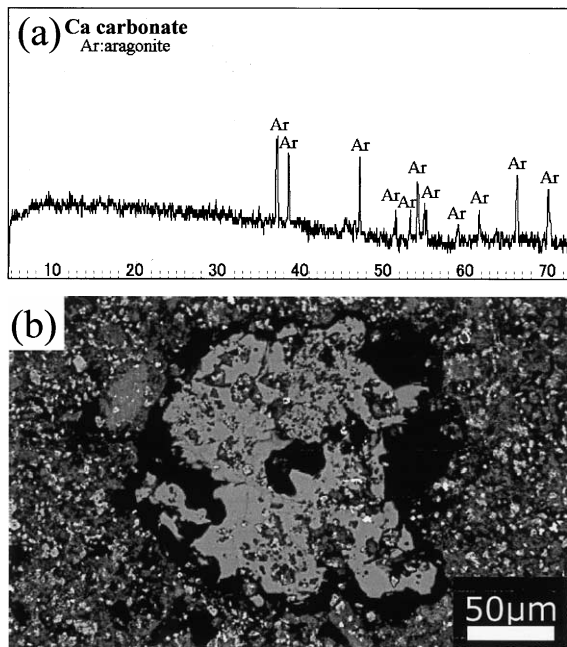


Fig. 4. Ca carbonate aragonite in matrix of the carbonate-rich lithology. (a) An X-ray diffraction pattern of aragonite that is obtained from a 20- μm piece of almost pure aragonite cut out from the matrix. (b) A BSE image of a large aragonite crystal, showing that it is intergrown with Mg-Fe carbonate and saponite.

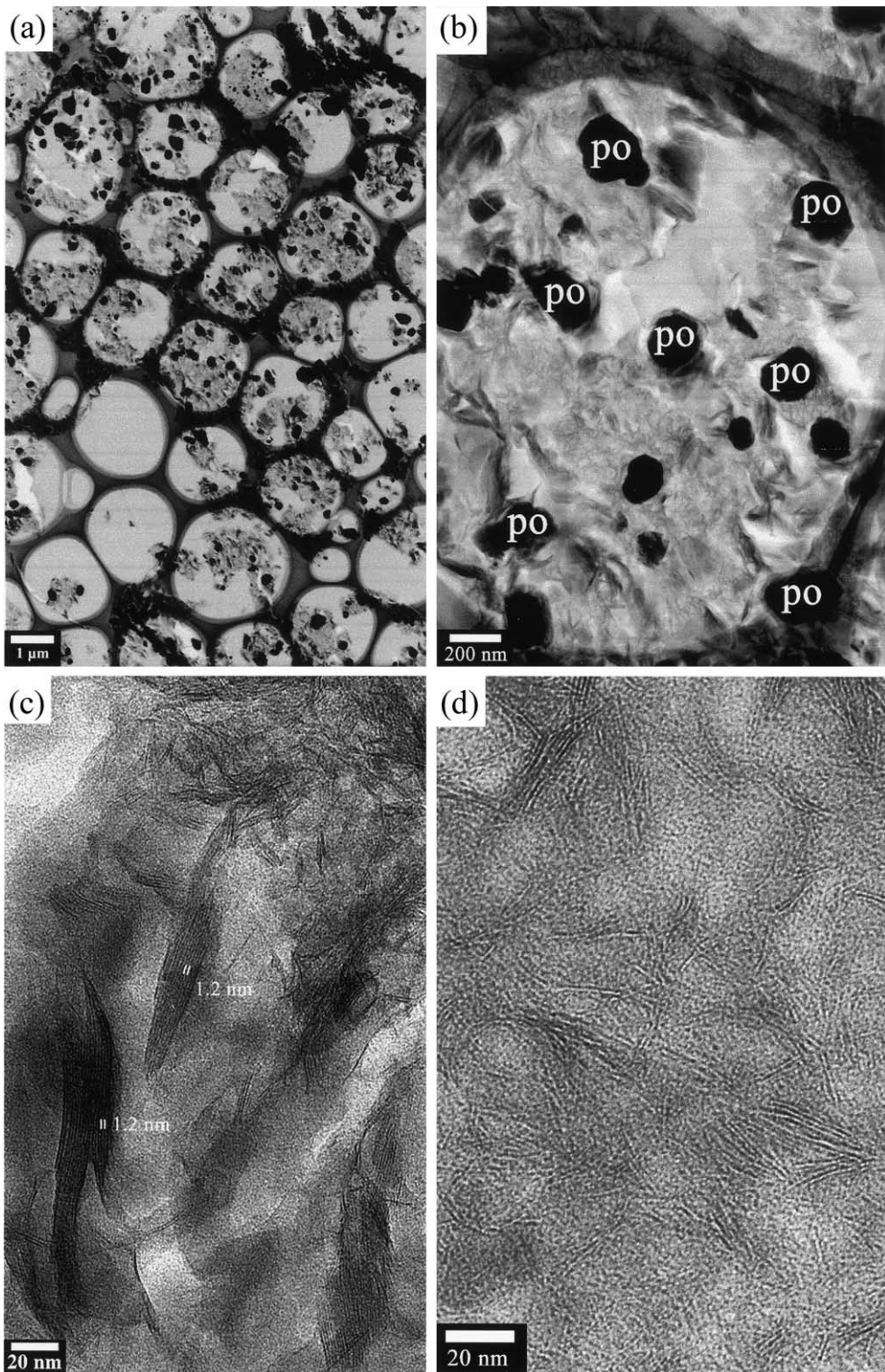
these phases (Fig. 3b). X-ray diffraction analysis confirmed the interpretation and revealed that both fine- and coarse-grained phyllosilicate are saponite (Fig. 3c). Saponite has a 001 basal spacing ranging from 13 to 14 Å and shows 02*l* and 20*l* prism reflections due to high degrees of stacking disorder. The diffraction analysis was performed on a 100- μm lump (Fig. 3a), and therefore the diffraction pattern (Fig. 3c) exhibits the averaged mineralogy of thousands of small mineral grains. The ferromagnesian carbonate is rich in siderite component judging from the peak center position of X-ray reflections, being consistent with

AEM analysis (Fig. 3b). The relative abundance of saponite and ferromagnesian carbonate is approximately 3:2, which was determined by applying the lever rule between the average compositions of matrix, phyllosilicates, and carbonates in Fig. 3b. The sulfide minerals pyrrhotite and pentlandite occur in matrix in lesser amounts (Fig. 3c). Rare ferroan olivine crystals and calcium carbonate are found in places of matrix. X-ray diffraction analysis showed that aragonite is the only calcium carbonate present (Fig. 4a,b). The aragonite grains were always intergrown with the Fe-Mg carbonates.

The unique matrix mineralogy of the carbonate-rich lithology of Tagish Lake can be clearly demonstrated in X-ray diffraction patterns in Fig. 3c, where the patterns of Orgueil CI (Fig. 3d) and matrix of Y-791198 CM2 (Fig. 3e) are also shown for comparison. In Tagish Lake matrix, saponite is the dominant phyllosilicate, while serpentine is the dominant phyllosilicate in CM2 matrix. CI is intermediate between the two, because it contains both saponite and serpentine in approximately equal quantities (Fig. 3d) – the comparable intensity of saponite 001 and serpentine 001 reflections in CI suggests that saponite/serpentine ratio is close to 1. Another difference to CI is the low abundance of magnetite and ferrihydrite in Tagish Lake matrix, instead pyrrhotite is abundant. Another difference to CM2 matrix is lack of tochilinite and mix-layered minerals of tochilinite and serpentine in Tagish Lake matrix. Carbonate mineralogy is also different: siderite, dolomite, and calcite are the main carbonate phases in the carbonate-rich lithology of Tagish Lake, CI, and CM2, respectively.

TEM investigation was performed on the three matrix lumps analyzed by X-ray diffraction. The matrix is a very porous material containing pyrrhotite, pentlandite, ferromagnesian carbonate,

Fig. 5. TEM photomicrographs of matrix of the carbonate-rich lithology. (a) A low-magnification bright field image. The image clearly shows the high porosity of the matrix. Honeycomb-like networks shown in (a), (b), and Fig. 8b are holey plastic films that support ultrathin sample sections. (b) An enlarged view of the matrix. Subhedral to euhedral pyrrhotite (po) crystals are embedded in the phyllosilicate dominant matrix. There are coarse- and fine-phyllosilicates. (c) A high-resolution image of the coarse-phyllosilicates (~ 20 nm thick). They have 1.2 nm basal lattice fringes. The spacing indicates that they are saponite. (d) A high-resolution image of the fine-phyllosilicates. The repetition of the basal lattice fringes is very poor. In general, the phyllosilicates have only several layers of basal fringes. Most of them have 1.2–1.3 nm interlayer spacing.



and magnetite embedded in phyllosilicates (Fig. 5a). Most sulfide grains are subhedral to euhedral pyrrhotite about 0.05–0.3 μm in diameter (Fig. 5b). Ferromagnesian carbonate occurs as discrete grains 0.2–3 μm across. Magnetite is a minor phase and cannot be detected by the X-ray diffraction analysis (Fig. 3c) and other Fe-bearing phases such as ferrihydrite and tochilinite were not identified. There are two types of phyllosilicates in the matrix. The first group occurs as irregularly shaped aggregates of coarse-grained phyllosilicates (Fig. 5c). Individual phyllosilicate crystallites in the aggregates are 100–200 \AA in width normal to their (001) planes. High-resolution TEM imaging and AEM analysis of these phyllosilicates show that they consist mainly of saponite (10.5–13.8 \AA basal spacing). Serpentine (7.0 \AA basal spacing) is very rare and is intergrown with saponite. The second group of phyllosilicates occur as aggregates of very fine-grained fibrous phyllosilicates (Fig. 5d). They are 10–50 \AA in width. Although their crystallinity is much lower than that of the coarse phyllosilicates, high-resolution TEM imaging and AEM analysis suggest that they consist mainly of saponite.

AEM analysis of matrix shows that the (Si+Al)/(Si+Al+Mg+Fe) elemental ratios of both fine- and coarse-grained phyllosilicates range from 0.5 to 0.6 (Fig. 3b). These compositions are quite unique, when compared with those in CI chondrites [9,10]. In Tagish Lake, coarse-grained phyllosilicates are more magnesian than fine ones. Mg/(Mg+Fe) ratios of the coarse-grained phyllosilicates are similar to those in the CI chondrites Orgueil and Ivuna [9,10]. Ferromagnesian carbonate in Tagish Lake ranges in composition from $(\text{Ca}_{0.05}\text{Mg}_{0.53}\text{Fe}_{0.42})\text{CO}_3$ to $(\text{Ca}_{0.15}\text{Mg}_{0.19}\text{Fe}_{0.66})\text{CO}_3$. The Mg-Fe carbonates show Mg/(Mg+Fe) ratios from 0.2 to 0.6 and CaO and MnO concentrations up to 10 and 2.0 wt%, respectively.

3.3. Clasts

Angular-shaped clasts variable in size from several to 800 μm occur in many places in matrix (Fig. 1). We identified 97 clasts with a longest dimension larger than 35 μm during a search of

60 mm^2 total area. 81 clasts are composed almost entirely of fine-grained material, whereas 16 clasts contain anhydrous mineral inclusions and/or chondrules. We found some clasts exhibiting especially insightful textures (Fig. 6a,b). The right portion of the clast in Fig. 6a contains some anhydrous mineral inclusions and an aggregate consisting of olivine and phyllosilicate that appears similar to a chondrule, while the left portion of the clast is a chondrule with a fine-grained rim and, interestingly, the upper right side of the rim is a part of the clast on the left. The texture clearly indicates that the chondrule rim and the clast are the same material. The clast in Fig. 6b also contains an irregular-shaped chondrule-like object. The lower portion of the clast is dominated by fine-grained material while the upper portion appears to be a chondrule with a fine-grained rim. Is this object in Fig. 6b 'a chondrule with rim' or 'a chondrule within a clast'? Many mineral inclusions and chondrules are off-centered in clasts having angular external morphology, which do not look like typical chondrules with fine-grained rim as one would observe in CM2 chondrites [11].

X-ray diffraction analysis was performed on seven 100- μm lumps of the fine-grained portions of clasts, three of which are from portions looking like fine-grained chondrule rim material, but again clasts are indistinguishable from chondrule rims texturally. The results showed that the mineralogy of the seven lumps is similar among the lumps, but differs from that of host matrix. The fine-grained portions of clasts consist of saponite, magnetite, Mg-Fe carbonate, pyrrhotite, and minor amounts of pentlandite (Fig. 7a). Compared with matrix (Fig. 3c), magnetite is very abundant and Mg-Fe carbonate is depleted in clasts. These mineralogical differences are reflected in chemical composition. The major-element compositions of clasts are plotted within a region defined by the following three components, Fe-apex (magnetite and pyrrhotite) and fine- and coarse-grained phyllosilicates (Fig. 7b), while those of matrix are a mixture of Fe-Mg carbonate and fine- and coarse-phyllosilicates (Fig. 3b).

Microtextures are also different between matrix and the fine-grained portions of clasts: the latter

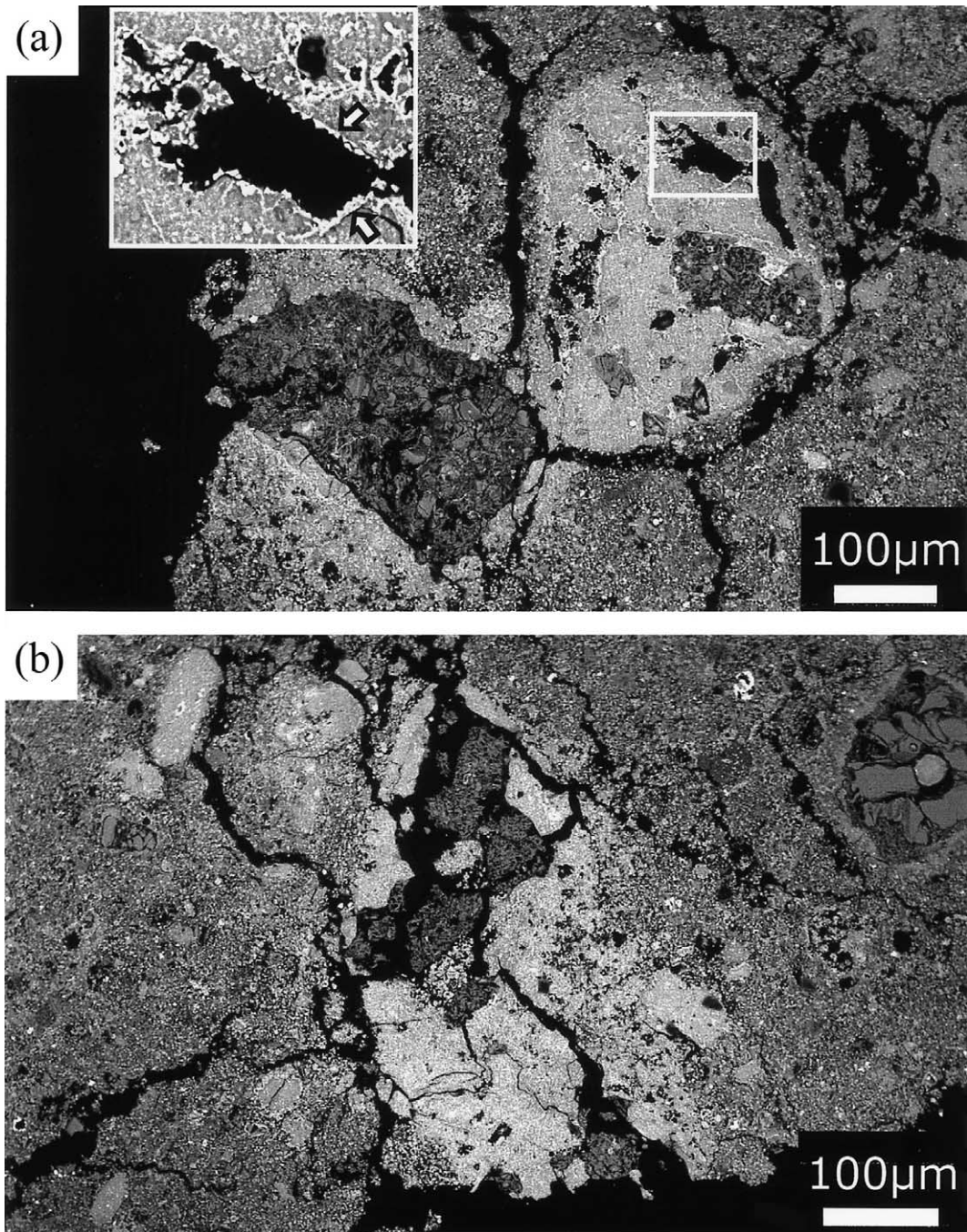


Fig. 6. BSE images of clasts occurring in the carbonate-rich lithology of Tagish Lake. (a) A clast consisting of two parts: a fine-grained part on the right contains an irregular-shaped chondrule-like object, silicate fragments, and large pores, and a coarse-grained part on the left is like a chondrule with rim. The two parts are connected. (Inset) An enlarged view of a large pore in the right part of the clast (framed in white). Small Mg-Fe carbonate crystals, indicated by arrows, are deposited on the walls of the pore. (b) A clast containing a chondrule. The lower side of the clast is dominated by fine-grained material.

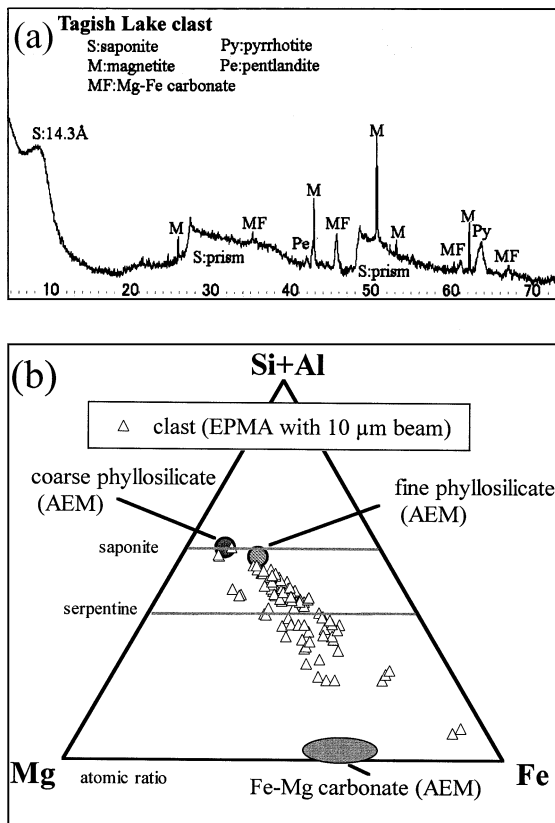


Fig. 7. (a) An X-ray diffraction pattern of a fine-grained portion of a clast. Saponite and magnetite are dominant phases, while Mg-Fe carbonate is a minor phase. (b) Chemical compositions of fine-grained portions of the clast. Eight clasts were measured by EPMA and three were measured by AEM, but the data were shown without distinction in the figure.

is much more compacted, less porous material than the former in BSE images (Fig. 8a). TEM observation also indicates that the clasts are compact material composed of phyllosilicates, magnetite, pyrrhotite, pentlandite, and ferromagnesian carbonate (Fig. 8b). The phyllosilicates have compositions and morphologies similar to those in matrix. They are coarse- and fine-grained phyllosilicates, both of which have compositions identical with those of matrix (Figs. 3b and 7b). Compared with matrix phyllosilicates, however, the fine-grained phyllosilicates are more abundant in clasts. In addition, intimate mixtures of serpentine

and saponite occur in places in clasts (Fig. 8c), but the abundance of the mixtures is sufficiently low that serpentine was not detected by X-ray diffraction (Fig. 7a). Magnetite frequently occurs as framboids (Fig. 8d). Pyrrhotite occasionally occurs as laths. Ferromagnesian carbonate is less abundant and smaller (typically 0.1–0.15 μm in diameter) than that in matrix. Isolated olivine grains occur in some places in clasts (Fig. 9a,b). They contain MnO up to 1 wt% and MnO content is equal to or higher than FeO (Fig. 9c), indicating that the olivine is LIME (low-iron-manganese-enriched: see [12]) olivine. LIME olivine is found in some IDPs and micrometeorites and is thought to be a direct condensate from the hot nebula [12,13]. Thin (~ 50 Å) saponite layers were observed on the surface of the LIME olivine grains (Fig. 9b).

4. Noble gases

The results of stepped heating analysis of a chip from the carbonate-rich lithology are shown in Table 1 for concentrations of all noble gases and isotopic ratios of He, Ne, and Ar. Kr and Xe isotopic ratios are given in the Appendix. Noble gases in the carbonate-rich lithology of Tagish Lake are dominated by a primordial component that was extracted mainly at 680, 910, and 1140°C in the stepped heating analysis (Table 1). The total concentration of ^{132}Xe , which is dominated by the primordial gas, is 1.6×10^{-8} cc/g (Table 1). This concentration is relatively high, but still within the range of CI and CM2 chondrites [14–16]. $^{36}\text{Ar}_{\text{trapped}}/^{132}\text{Xe}$ and $^{84}\text{Kr}/^{132}\text{Xe}$ ratios are 83 and 0.84, respectively, which are close to those of Xe-Q [17–19], indicating that Q gas is a major component in heavy noble gases. It has recently been proposed that fullerenes may be a major carrier of Q gas in Tagish Lake meteorite [20], although this report has not been verified. Xe isotopic ratios (Appendix) indicate that Xe is a mixture of Xe-Q, -HL, -S, and atmospheric Xe. Contributions from Xe-HL are pronounced at 910°C where both $^{134}\text{Xe}/^{132}\text{Xe}$ and $^{136}\text{Xe}/^{132}\text{Xe}$ ratios ($^{134}\text{Xe}/^{132}\text{Xe} = 0.397$ and $^{136}\text{Xe}/^{132}\text{Xe} = 0.332$) are much higher than those of Xe-Q ($^{134}\text{Xe}/^{132}\text{Xe} =$

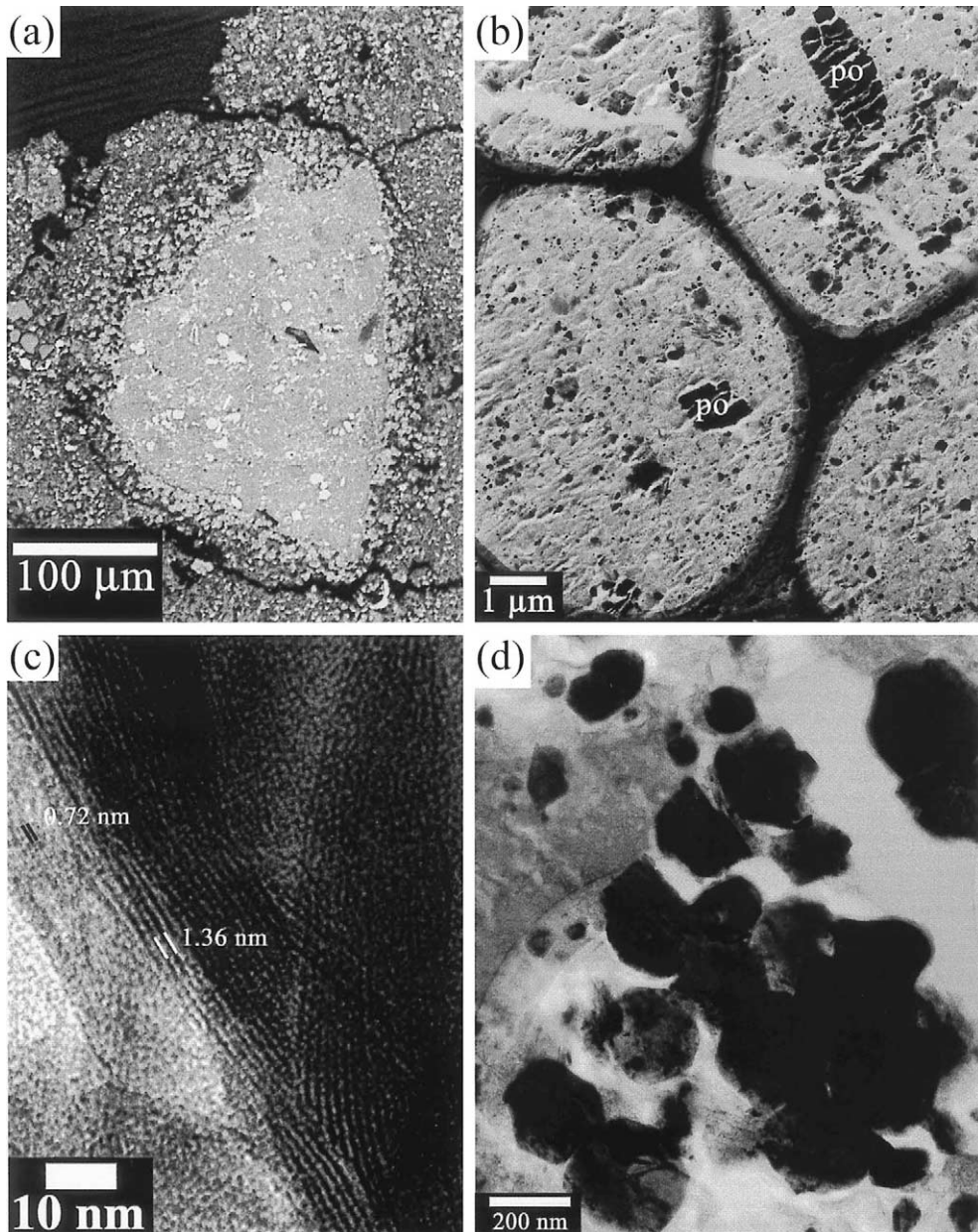
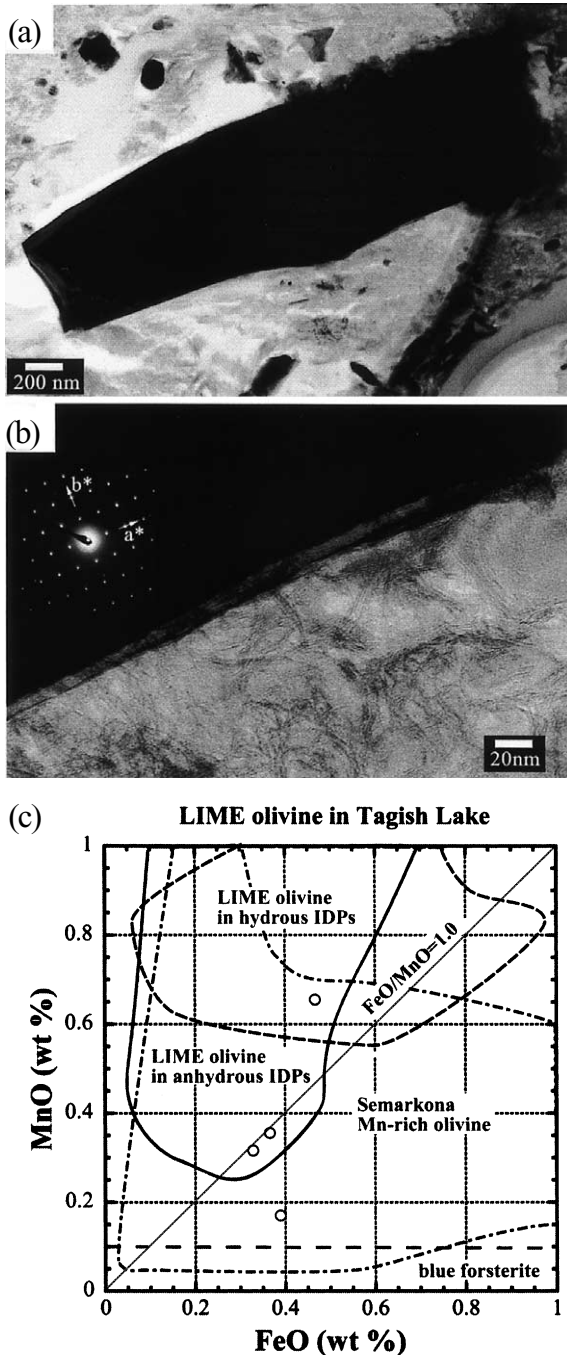


Fig. 8. SEM and TEM photomicrographs of a clast in the carbonate-rich lithology. (a) A BSE image of a clast, showing that it is a compact material compared with matrix surrounding it. Fine-grained (< 5 μm across) Mg-Fe carbonate crystals are rimming the clast. (b) A low-magnification BF image of a clast, showing that the clast is much more compact than the matrix (see Fig. 5a) in TEM images. Some large (> 1 μm across) pyrrhotite (po) crystals are present. (c) A high-resolution image of the fine-phyllosilicates, showing that a small amount of serpentine (0.72-nm basal fringes) coexists with saponite (1.36-nm basal fringes) in the clast. (d) A BF image of framboidal aggregates of small euhedral to subhedral magnetite crystals (< 300 nm across).

0.381 and $^{136}\text{Xe}/^{132}\text{Xe} = 0.319$ [17]). Xe-HL contribution to the total Xe concentration was calculated to be 1.9% based on $^{136}\text{Xe}/^{132}\text{Xe}$ ratios of 910, 1140, 1400, and 1700°C fractions.



Solar-type noble gases are absent from the carbonate-rich lithology, judging from the elemental and isotopic ratios. The $(^{20}\text{Ne}/^{36}\text{Ar})_{\text{trap}}$ ratio is 0.3, which is in the range of primordial values, but much lower than typical ratios (10–30) observed in solar-gas-rich meteorites. Ne isotopic ratios indicate that Ne is a mixture of primordial (Ne-A2 and -E) and cosmogenic components, except for Ne extracted at 500°C whose datum plots above the tie line connecting Ne-A2 and cosmogenic components in the $^{20}\text{Ne}/^{22}\text{Ne}$ – $^{21}\text{Ne}/^{22}\text{Ne}$ diagram (Fig. 10a). At face value, the high $^{20}\text{Ne}/^{22}\text{Ne}$ ratio of this step thus appears to indicate the presence of solar gases, but this possibility is again excluded by the very low $(^{20}\text{Ne}/^{36}\text{Ar})_{\text{pri}}$ ratio of this step of 0.1. Therefore, the high $^{20}\text{Ne}/^{22}\text{Ne}$ ratio of this step is likely due to contributions of Ne-Q ($^{20}\text{Ne}/^{22}\text{Ne} = 10.7$ [17,18]). The results of Ne analysis of a slice of the carbonate-rich lithology (Table 2 and Fig. 10b) are similar to those of the chip. Total ^{20}Ne concentrations and isotopic ratios ($^{20}\text{Ne}/^{22}\text{Ne}$, $^{21}\text{Ne}/^{22}\text{Ne}$) of the slice are 4.17×10^{-7} cc/g and (6.40, 0.175), respectively, which are close to those of the chip of 4.08×10^{-7} cc/g and (6.23, 0.205), respectively. Therefore, Ne in the slice is also a mixture of primordial and cosmogenic components without any solar-gas contribution. The cosmic-ray exposure age cannot be determined precisely, because the shielding depth of our samples in the Tagish Lake meteoroid is unknown. Using Na, Mg, Al, Si, Ca, Fe, and Ni concentrations in Tagish Lake

Fig. 9. A LIME olivine crystal in a clast in the carbonate-rich lithology. Clasts in Tagish Lake contain a small amount of anhedral to subhedral LIME olivine crystals. (a) A BF image of a large ($2.6 \times 0.8 \mu\text{m}$) LIME olivine. (b) An enlarged BF image of the crystal shown in (a). A thin layer (about 10 nm thick) of phyllosilicate is observed on the surface of the crystal. (c) MnO and FeO concentrations of the LIME olivine crystals (open circles), obtained by AEM analysis. Fe-Mg zoning was not obvious in LIME olivines, thus the data in the figure represent compositions of bulk olivine crystals. Compositional ranges of LIME olivines in IDPs, those in the matrix of Semarkona LL3.0 chondrite, and those of forsterite with blue cathode luminescence in chondrites are also shown for comparison (data from [12,13]). Compositions LIME olivines in Tagish Lake are in the range of those in anhydrous IDPs and Semarkona chondrite.

Table 1
Noble-gas compositions of a chip (18.9 mg) from the carbonate-rich lithology of Tagish Lake

	⁴ He	³ He/ ⁴ He	²⁰ Ne	²⁰ Ne/ ²² Ne	²¹ Ne/ ²² Ne	²² Ne-Q ^a	²² Ne-A2	²² Ne-C ^b	²² Ne-E	⁴⁰ Ar	³⁸ Ar/ ³⁶ Ar	⁴⁰ Ar/ ³⁶ Ar	⁸⁴ Kr	¹³² Xe
260°C	33.6	0.00034 ± 0.0006	0.50	7.74 ± 0.72	0.067 ± 0.012	0.01 ± 0.001	0.05 ± 0.01	0	0	143	0.1898 ± 0.0035	62.5 ± 4.9	0.11	0.32
500°C	218	0.00026 ± 0.00001	3.18	9.12 ± 0.21	0.098 ± 0.005	0.07 ± 0.03	0.28 ± 0.04	0	0	476	0.1875 ± 0.0012	33.3 ± 0.8	0.36	0.84
680°C	7500	0.00428 ± 0.00005	67.5	4.30 ± 0.03	0.498 ± 0.007	1.12 ± 0.58	5.78 ± 0.73	8.30 ± 0.48	0.55 ± 0.41	4870	0.1934 ± 0.0007	20.3 ± 0.1	2.32	1.84
910°C	51700	0.00018 ± 0.00001	303	712 ± 0.04	0.097 ± 0.001	2.56 ± 1.33	32.13 ± 1.68	3.18 ± 0.19	4.70 ± 0.37	1340	0.1889 ± 0.0004	2.44 ± 0.02	6.51	7.72
1140°C	2060	0.00031 ± 0.00001	26.0	4.88 ± 0.05	0.134 ± 0.003	1.47 ± 0.76	1.15 ± 0.96	0.69 ± 0.04	2.02 ± 0.19	102	0.1883 ± 0.0003	0.3245 ± 0.003	2.64	3.36
1400°C	225	0.00059 ± 0.00003	7.87	5.34 ± 0.07	0.462 ± 0.011	0.73 ± 0.38	0.31 ± 0.01	0.56 ± 0.29	0.09 ± 0.09	74	0.1882 ± 0.0004	0.474 ± 0.001	1.25	1.69
1700°C	1.70	0.00092 ± 0.00021	n.d. ^c	–	–	n.d.	n.d.	n.d.	n.d.	47	0.1899 ± 0.0042	28.0 ± 6.9	0.02	0.04
Total	61700	0.00068	408	6.23	0.204	6.0 ± 3.1	39.7 ± 3.6	12.7 ± 0.8	7.4 ± 1.0	7050	0.1898	5.53	13.2	15.8

Concentrations are given in the unit of 10^{-9} cc STP/g. Errors of concentrations are 5% for ⁴He, ²⁰Ne, ⁴⁰Ar and 10% for ⁸⁴Kr and ¹³²Xe. Errors of isotopic ratios include mass discrimination uncertainties.

^a ²²Ne-Q concentrations are obtained from primordial ³⁶Ar concentrations, (²⁰Ne/²²Ne)_Q ratios.

^b ²²Ne-C means cosmogenic ²²Ne. The concentrations vary with (²⁰Ne/²²Ne, ²¹Ne/²²Ne)_C ratios. Mean values and ranges are given.

^c n.d. means not detected.

[1] and Ne production rates from these elements at 2 to 100 cm depth [21], the depth with the highest ²¹Ne production rate was obtained to be 21 cm. At this depth, ²¹Ne production rate is 1.94×10^{-9} cc STP/g Ma and (²⁰Ne/²²Ne, ²¹Ne/²²Ne) ratio is 0.771, 0.925, respectively. In this calculation, the radius of the Tagish Lake meteoroid was assumed to be 120 cm, because the size of the meteoroid before atmospheric entry was large (~ 3 m radius [1]). The ²¹Ne production rates at shallower or deeper than 21 cm are small down to 1.29×10^{-9} cc STP/g Ma. Using the highest ²¹Ne production rates at 21 cm, the minimum exposure age was determined to be 5.0 and 6.0 Myr for the slice and the chip, respectively. The chip and slice were irradiated in close proximity in the meteoroid, because both are from single large fragment of Tagish Lake. Thus, we defined 5.5 ± 0.7 Myr as the minimum exposure age. The true exposure age must be longer than that.

5. Discussion and conclusions

5.1. Pristine features found in Tagish Lake

Concentrations of primordial noble gases in the

carbonate-rich lithology of Tagish Lake meteorite are among the highest ones observed in carbonaceous chondrites, indicating that Tagish Lake contains very pristine solar system material. The high concentration of Q-gas suggests that Tagish Lake as a whole has never experienced severe thermal processing, similar to primitive carbonaceous chondrites such as CI and CM. The low (³⁶Ar)_{trap}/¹³²Xe ratio (= 83) of the Tagish Lake sample indicates no detectable contribution of Ar-rich noble gases that are ubiquitously present in *anhydrous* primitive chondrites [22,23]. The depletion of Ar-rich gases is a common feature of *hydrous* carbonaceous chondrites [23], and is consistent with the aqueously altered nature of Tagish Lake. On the other hand, our analysis revealed that noble gases located in various presolar grains are enriched in Tagish Lake relative to other carbonaceous chondrites. In order to estimate the abundances of presolar grains, the concentrations of ²²Ne-E and ²²Ne-A2, which are known to be located in interstellar SiC/graphite and diamond, respectively, were calculated based on Ne concentrations and isotopic ratios. In the calculations, Ne of each temperature step in stepwise heating analysis was separated into the four components, Ne-A2, -E, -Q, and -cosmogen-

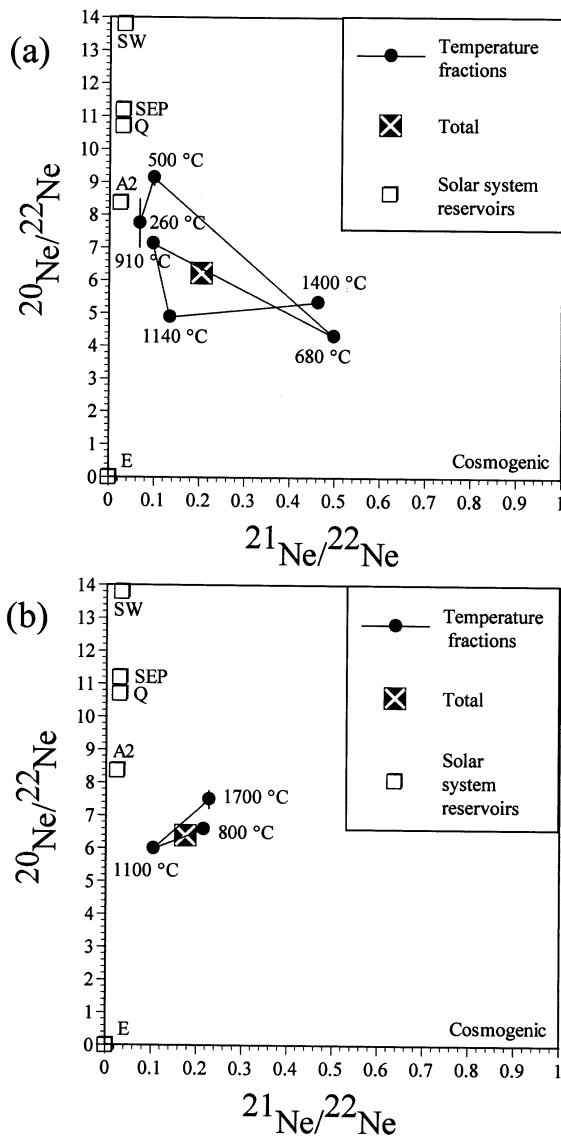


Fig. 10. (a) Ne isotopic ratios of a chip from the carbonate-rich lithology of Tagish Lake meteorite. A major portion of Ne was extracted in 680, 910, and 1140 °C fractions (Table 1). From the diagram, it is indicated that 910 °C fraction is enriched in Ne-A2 and 1140 °C fraction is enriched in Ne-E. (b) Ne isotopic ratios of a slice of the carbonate-rich lithology. Ne in 1100 °C fraction is enriched in Ne-E.

ic (Tables 1 and 2). At first, ^{20}Ne -, ^{21}Ne -, ^{22}Ne -Q concentrations were determined from primordial ^{36}Ar concentrations, ($^{20}\text{Ne}/^{36}\text{Ar}$)_Q ratio (0.05 ± 0.02 given in [19]), and ($^{20}\text{Ne}/^{22}\text{Ne}$, $^{21}\text{Ne}/^{22}\text{Ne}$)_Q ratio ((10.7, 0.0294), respectively, given in [18]).

Then the obtained Ne-Q concentrations were subtracted from the measured Ne concentrations, and the remaining Ne was separated into Ne-A2, -E, and -cosmogenic components. For the component separation, the following isotopic ratios were used: ($^{20}\text{Ne}/^{22}\text{Ne}$, $^{21}\text{Ne}/^{22}\text{Ne}$)_{A2} = (8.500, 0.036) [24] and ($^{20}\text{Ne}/^{22}\text{Ne}$, $^{21}\text{Ne}/^{22}\text{Ne}$)_E = (0, 0). Cosmogenic Ne isotopic ratios change with shielding depth, thus the possible ranges of ($^{20}\text{Ne}/^{22}\text{Ne}$, $^{21}\text{Ne}/^{22}\text{Ne}$)_C = (0.757 ~ 0.779, 0.869 ~ 0.946), respectively, were obtained from ^{20}Ne -, ^{21}Ne -, and ^{22}Ne production rates at various shielding depths, in the same way as shown in the result in Section 4.

The results show that ^{22}Ne -E and ^{22}Ne -A2 concentrations in the meteorite chip are $7.4 \pm 1.0 \times 10^{-9}$ cc/g and $4.0 \pm 0.4 \times 10^{-8}$ cc/g, respectively (Table 1) and those in the meteorite slice are $8.2 \pm 1.1 \times 10^{-9}$ cc/g and $3.9 \pm 0.5 \times 10^{-8}$ cc/g, respectively (Table 2). The ^{22}Ne -E and ^{22}Ne -A2 concentrations in both samples are at the maximum level observed for CI and CM chondrites (Fig. 11). These results require high abundances of presolar grains in the Tagish Lake samples and this conclusion is consistent with the signatures of carbon and nitrogen isotopes in this meteorite [25].

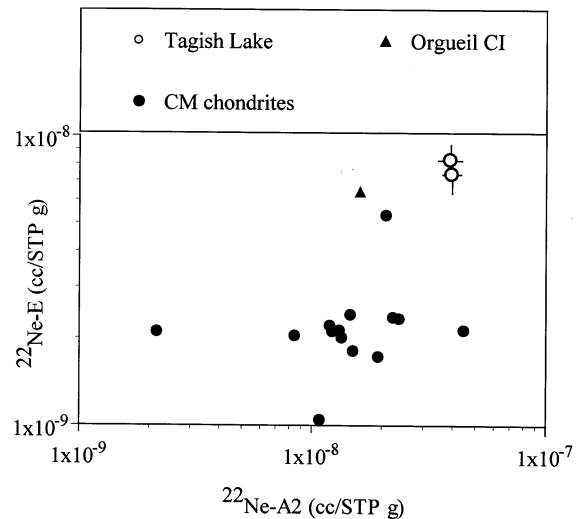


Fig. 11. A diagram showing concentrations of ^{22}Ne -A2 and ^{22}Ne -E in the two samples from the carbonate-rich lithology of Tagish Lake, those in Orgueil CI, and those in CM chondrites. CM data are from [35,36] and CI is from [37].

Table 2
Ne and Ar compositions of a slice (3.5 mg) from the carbonate-rich lithology of Tagish Lake

	²⁰ Ne	²⁰ Ne/ ²² Ne	²¹ Ne/ ²² Ne	²² Ne-Q ^a	²² Ne-A2	²² Ne-C ^b	²² Ne-E	⁴⁰ Ar	⁴⁰ Ar/ ³⁶ Ar	³⁸ Ar/ ³⁶ Ar
800°C	254	6.58 ± 0.07	0.215 ± 0.008	2.88 ± 1.50	25.53 ± 1.89	8.00 ± 0.46	2.17 ± 0.57	4290	6.95 ± 0.02	0.1920 ± 0.001
1100°C	143	5.99 ± 0.13	0.104 ± 0.006	2.94 ± 1.53	12.93 ± 1.93	2.10 ± 0.12	5.89 ± 0.39	368	0.584 ± 0.002	0.1892 ± 0.001
1700°C	19.6	7.49 ± 0.26	0.227 ± 0.003	1.56 ± 0.81	0.68 ± 0.65	0.42 ± 0.36	0.18 ± 0.20	1490	4.47 ± 0.01	0.1894 ± 0.001
Total	417	6.40	0.175	7.4 ± 3.8	39.1 ± 4.5	10.5 ± 0.7	8.2 ± 1.1	6150	3.891	0.1904

Concentrations are given in the unit of 10⁻⁹ cc STP/g. Errors of concentrations are 5% for ⁴He, ²⁰Ne, ⁴⁰Ar and 10% for ⁸⁴Kr and ¹³²Xe. Errors of isotopic ratios include mass discrimination uncertainties.

^a ²²Ne-Q concentrations are obtained from primordial ³⁶Ar concentrations, (²⁰Ne/³⁶Ar)_Q, and (²⁰Ne/²²Ne)_Q ratios.

^b ²²Ne-C means cosmogenic ²²Ne. The concentrations vary with (²⁰Ne/²²Ne, ²¹Ne/²²Ne)_C ratios. Mean values and ranges are given.

5.2. Tagish Lake as an accretionary breccia

The carbonate-rich lithology is dominated by matrix material that is a loosely packed, porous aggregate consisting mainly of fine saponite and ferromagenesian-carbonate particles (Fig. 3a). The two minerals are evenly distributed throughout the matrix (Fig. 3a). This texture indicates that matrix experienced no dynamic compression or deformation after the formation of the carbonate-rich lithology. The matrix contains many inclusions, most of which are fine-grained clasts with angular morphology (Fig. 1). Our analysis showed that the clasts and the host matrix are different in texture, mineralogy, and bulk chemical compositions. The clasts exhibit a massive, compact appearance with porosity much lower than matrix (Fig. 8a). Clasts are rich in magnetite and saponite–serpentine mixtures and poor in carbonates relative to the host matrix. Broad beam microprobe analysis showed that the chemical compositions differ between the clast and the host matrix (Figs. 3b and 7b). All observations indicate that the carbonate-rich lithology is a breccia, consisting mainly of two different fine-grained phyllosilicate-rich materials.

The compositions of light noble gases indicate that solar noble gases are absent from the carbonate-rich lithology. The absence of solar noble gases suggests that the carbonate-rich lithology was formed in an environment where solar wind was shielded. There are two cases for breccia formation under solar-wind-shielded conditions: (1) formation by mechanical mixing at deep inside of a planetesimal and (2) formation by accretion of various objects at surface layers of a planetesimal

located within the massive dust–gas cloud in the early solar nebula. The lack of solar noble gases precludes breccia formation at planetesimal surfaces after nebular dissipation (i.e., regolith breccias), because this would have resulted in an enrichment of solar-wind noble gases in the constituent material of the breccia, as is often observed in other meteorites (e.g. [16,26]). In case (1), the clasts and the matrix should have been mechanically mixed by impacts at some depth in a planetesimal to form the carbonate-rich lithology. But such dynamic effects are absent in the matrix, ruling out the possibility of case (1). On the other hand, case (2) is consistent with the signatures of the texture of the carbonate-rich lithology. Fine-grained porous matrix is dominant and it contains many clasts, which can be formed by gentle accretion of fine particles together with the clasts. Therefore, the most probable mechanism for the formation of carbonate-rich lithology is simultaneous accretion of clasts, chondrules, and matrix material to the Tagish Lake parent asteroid in a massive dust cloud from which solar wind was shielded. Such a breccia is classified as an accretionary breccia [27].

The clasts often enclose chondrules and anhydrous silicates such as LIME olivines. This suggests that the clasts have been a part of earlier-formed chondritic materials and from which they were excavated and incorporated into new locations. The highly compacted, angular morphology of the clasts suggests that they have experienced compression and fragmentation on the earlier-formed chondritic bodies due to impacts. The impact must have been very weak, because saponite in the clasts is not dehydrated (as would be re-

vealed by a reduction of the (001) basal spacing). Experimental heating of saponite in Tagish Lake showed that onset of dehydration is 500°C [8]. This indicates that the clasts never been heated to that temperature, which is consistent with the types of sulfides present. In addition, mild impacts reduce the porosity of the matrix without melting of material, as is observed in some shocked CV3 chondrites [28]. X-ray diffraction analysis showed that the mineralogy is similar among the Tagish Lake clasts, suggesting that all the clasts may originate from a single earlier-formed chondritic body. There is a possibility that the earlier-formed body is the Tagish Lake parent body itself, because the mineralogy of the clasts is similar to that of matrix material in *carbonate-poor* lithology [2]. More mineralogical comparison is needed to solve this issue. In this study we showed that the carbonate-rich lithology is an accretionary breccia that contains clasts formed by disagglomeration an earlier-formed body. It is thus suggested that, in the course of planetesimal formation, agglomeration and disagglomeration occurred repeatedly in a dense dust cloud where solar wind from the infant sun did not reach. This suggestion has a major bearing on whether asteroids today are mainly loosely bound rubble piles.

5.3. 'Chondrule within clast' or 'chondrule with rim'?

Tomeoka et al. [29] found that small, fine-grained phyllosilicate-rich clasts and phyllosilicate-rich chondrule rims in the Vigarano CV3 chondrite are similar in mineralogy and concluded that the chondrule rims in this meteorite were remnants of the clasts around chondrules. In this study, we found similar features in the carbonate-rich lithology of Tagish Lake and further discovered direct textural evidence demonstrating that fine-grained clasts and fine-grained chondrule rims are the same material (Fig. 6a). Chondrules are off-centered in angular-shaped clasts (Fig. 6b), thus 'chondrule within clast' and 'chondrule with rim' are indistinguishable. Therefore, objects which appear like chondrule rim material can be recognized to be a remnant of fine-grained clasts around chondrules. Our results suggest that

Appendix. Kr and Xe isotopic ratios of the carbonate-rich lithology of Tagish Lake

	78/84	80/84	82/84	83/84	84/84	86/84	124/132	126/132	128/132	129/132	130/132	131/132	132/132	134/132	136/132
260°C	0.0060 ±0.0005	0.040 ±0.002	0.199 ±0.005	0.193 ±0.008	1	0.308 ±0.011	0.0036 ±0.0001	0.0035 ±0.0002	0.073 ±0.001	0.989 ±0.010	0.154 ±0.003	0.795 ±0.006	1	0.386 ±0.005	0.322 ±0.004
500°C	0.0064 ±0.0004	0.040 ±0.001	0.204 ±0.004	0.197 ±0.002	1	0.306 ±0.002	0.0037 ±0.0002	0.0034 ±0.0002	0.074 ±0.001	0.996 ±0.005	0.155 ±0.002	0.798 ±0.005	1	0.386 ±0.003	0.323 ±0.002
680°C	0.0065 ±0.0003	0.041 ±0.001	0.205 ±0.001	0.202 ±0.001	1	0.315 ±0.001	0.0047 ±0.0001	0.0042 ±0.0001	0.083 ±0.001	1.054 ±0.004	0.165 ±0.001	0.822 ±0.009	1	0.380 ±0.002	0.313 ±0.002
910°C	0.0063 ±0.0002	0.040 ±0.001	0.204 ±0.002	0.200 ±0.001	1	0.315 ±0.002	0.0049 ±0.0001	0.0044 ±0.0001	0.086 ±0.001	1.032 ±0.002	0.170 ±0.001	0.830 ±0.001	1	0.397 ±0.002	0.332 ±0.001
1140°C	0.0064 ±0.0002	0.040 ±0.001	0.205 ±0.001	0.200 ±0.002	1	0.313 ±0.003	0.0047 ±0.0001	0.0042 ±0.0001	0.085 ±0.001	1.030 ±0.002	0.168 ±0.001	0.825 ±0.003	1	0.388 ±0.002	0.320 ±0.001
1400°C	0.0064 ±0.0002	0.041 ±0.001	0.204 ±0.002	0.199 ±0.002	1	0.313 ±0.003	0.0047 ±0.0002	0.0041 ±0.0001	0.084 ±0.001	1.032 ±0.002	0.167 ±0.001	0.827 ±0.003	1	0.386 ±0.002	0.320 ±0.002
1700°C	0.0077 ±0.0046	0.042 ±0.004	0.199 ±0.018	0.188 ±0.010	1	0.316 ±0.010	0.0045 ±0.0015	0.0036 ±0.0015	0.081 ±0.004	1.046 ±0.012	0.162 ±0.004	0.832 ±0.019	1	0.385 ±0.013	0.323 ±0.013

‘chondrule rims’ in the carbonate-rich lithology were formed in the earlier-formed clasts’ parental material, which is consistent with the rim formation model proposed by [29]. Unlike Tagish Lake, Vigarano contains large amounts of solar noble gases [14], thus it is a regolith breccia consisting of a variety of material with different origins. Therefore, although the same mechanism seems to have worked for the rim formation in Vigarano and Tagish Lake, subsequent processes must have differed between them.

In CM chondrites, most chondrules and inclusions have fine-grained rims that are believed to have formed by accretion of dust onto chondrules [11,30–32]. Rims in CM chondrites differ in many respects from Tagish Lake rims. The former have a rounded shape and are continuous all around the enclosing chondrule, while the latter have an angular shape and are discontinuous (Fig. 6b). In addition, fine-grained angular clasts, as are observed in both Tagish Lake and Vigarano, were not found in CM chondrites. Therefore, CM rims lack direct evidence suggestive of a remnant of fine-grained clasts around chondrules.

5.4. Features indicative of aqueous alteration

The carbonate-rich lithology exhibits many mineralogical features indicative of extensive aqueous alteration. The degree of alteration varies between chondrules, but there are many chondrules that retain anhydrous silicates. This means that Tagish Lake is a petrologic type-2 carbonaceous chondrite, supporting the classification given by [2]. The matrix mineralogy of Tagish Lake is far different from that of CM and CI chondrites (Fig. 3c–e). During aqueous alteration abundant Mg-Fe carbonates were formed in Tagish Lake. Formation of the Mg-Fe carbonates postdates the formation of clasts and matrix, because the carbonates form on the surfaces of the clasts (Fig. 7a; also see [2]). We found textural evidence demonstrating that ferromagnesian carbonate is a late-stage precipitate product. Small euhedral carbonates cover the inner wall of large pores within the fine-grained clasts (Fig. 6a, inset), which indicates that the carbonate was precipitated from solutions having filled or passed

through the pores. The precipitation of ferromagnesian carbonate in both clasts and matrix must have taken place simultaneously, because AEM analysis showed that the carbonate in clasts and matrix has the same compositions. The large pores within the fine-grained clasts (Fig. 6a, inset) may be pseudomorphs of preexisting solid volatiles such as H₂O or CO₂ ice, because they have flat-plane walls. Calcium carbonate aragonite was present in some places in matrix. Aragonite is stable in solutions that are warm (> 50°C) [33] and rich in cations such as magnesium [34]. The solutions altering the carbonate-rich lithology during aragonite deposition may have had such properties.

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