

Yoshiokaite, a new Ca,Al-silicate mineral from the Moon

DAVID T. VANIMAN, DAVID L. BISH

Geology and Geochemistry Group, MS D462, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, U.S.A.

ABSTRACT

Yoshiokaite ($\text{Ca}_{8-(x/2)}\square_{(x/2)}\text{Al}_{16-x}\text{Si}_x\text{O}_{32}$) is hexagonal with a nepheline-like structure. It is a metastable phase formed by devitrification of glass at $\sim 950\text{--}1200^\circ\text{C}$ in regions of the system $\text{CaO-Al}_2\text{O}_3\text{-SiO}_2$ where corundum or hibonite are the stable liquidus phases. Yoshiokaite occurs as shocked crystal fragments and in devitrified glasses from one part of a rare Apollo 14 regolith breccia. Chemical systematics of yoshiokaite support an origin by volatilization of silica in impact melts from an almost pure anorthositic lunar highlands terrain. The severe impact conditions for formation, the requirement of an anorthositic impact target, and indirect evidence of great age make yoshiokaite a mineral characteristic of the ancient lunar highlands.

INTRODUCTION

Although the most spectacular samples from the Moon are large rocks, it is the small fragments from the lunar regolith that provide the broadest sampling of the Moon. Over four billion years of meteorite impacts have distributed small and varied rock fragments across the lunar surface. In the absence of thorough Moon-wide sampling, the Moon is still being explored through proxy analysis of these small and often exotic samples in its surface debris.

There has been much recent interest in regolith breccias, coherent pieces of regolith formed by impact compression and heating (Schaal and Hörz, 1980). Regolith breccias range from poorly coherent to tough and from porous to dense, depending on the magnitude of shock and heat effects involved in their formation. However, they all contain remnants of original soil particles. Some regolith breccias preserve soil features that may be about four billion years old (McKay et al., 1986). At that time the major cataclysmic impacts on the Moon were subsiding and smaller-scale but pervasive impact reworking began to create the present-day lunar regolith. Ancient regolith breccias thus provide a glimpse of an older lunar surface.

OCCURRENCE, CLASSIFICATION, AND NAME

Apollo 14 was the last of the Apollo landing sites to be traversed only by astronauts on foot. Without the motorized rover, their traverse range was limited to a radius of about 1.3 km. At a distance of about 224 m from their lander, in the bottom of a 30-cm-deep trench, they collected the two-gram rock that is regolith breccia 14076. Initial descriptions of sample 14076 led to the recognition of two distinct parts to this sample, separated by a sharp contact (this description is summarized by Jerde et al., 1990). Further work by Warren et al. (1987) and Jerde et al. (1990) confirmed that one part of sample 14076

is a regolith breccia typical of the local Apollo 14 regolith, whereas the other part (represented in sample split 14076,1) is exotic. The exotic part of this sample is very high in Al and has an extremely small maturity index I_s/FeO (the ratio of fine-grained Fe metal to FeO), which increases with impact-induced reduction of soil FeO by solar wind gases (H, C, N). The I_s/FeO ratio is 0.03 in the exotic portion of 14076, smaller than the value for most other regolith breccias (generally >0.2) and much smaller than that for soils (generally 20 to 90). The data for this portion of sample 14076 suggest that it is from an old and previously unknown regolith.

Compositions of glass and devitrified-glass fragments in sample 14076 and several other Apollo 14 regolith breccias were studied by Vaniman et al. (1988). One population of devitrified glass is exceptionally Al-rich and Si-poor and seems to account for the anomalously Al-rich nature of 14076,1. This devitrified glass is similar to glasses found elsewhere on the Moon but only rarely at Apollo 14 ($\sim 1/850$ of the glasses of Brown et al., 1971; $\sim 8/1026$ of the glasses of Finkelman, 1973; $\sim 1/400$ of the glasses of Simon et al., 1982). These glasses generally occur as small (<0.2 mm) colorless spheres; their high-Al, Si-poor composition has led to the acronym "HASP" (Naney et al., 1976). The HASP-like portions of sample 14076, however, are devitrified—something which has not been found in other HASP samples. This difference is significant because the anticipated silicate devitrification products are known minerals such as anorthite, corundum, and gehlenite (Vaniman et al., 1988). Whereas anorthite is common throughout the lunar highlands, the only suggested occurrence of gehlenite (tentative description in Masson et al., 1972) and one of the few occurrences of corundum (Christophe-Michel-Levy et al., 1972) are from Apollo 14. It was at first considered that the devitrified HASP-like fragments in sample 14076 could be fine-grained mixtures of these three phases (Vaniman et al., 1988). This hypothesis was rejected, however, when

several single crystals with the HASP-like composition were found in this regolith breccia (Vaniman et al., 1989). The most silica-poor fragments have a stoichiometry near $\text{Ca}_3\text{Al}_6\text{Si}_2\text{O}_{16}$, and the full range of compositions overlaps that of a metastable phase made by devitrification of synthetic $\text{CaO-Al}_2\text{O}_3\text{-SiO}_2$ glasses, at temperatures of about 950–1200 °C (Yoshioka, 1970a). The metastable phase that Yoshioka synthesized was a solid solution between $\text{CaAl}_2\text{Si}_2\text{O}_8$ and CaAl_2O_4 , with a nepheline-like structure and formula $\text{Ca}_{8-(x/2)}\square_{(x/2)}\text{Al}_{16-x}\text{Si}_x\text{O}_{32}$. Yoshioka's metastable solid solution has properties consistent with those of the mineral we found in regolith breccia 14076. The characterization and even the discovery of this new mineral from the Moon owes much to the synthesis studies by Yoshioka (1935–1983). Because he carried out the basic research on the synthetic phase, we have named the new mineral yoshiokaite. The new mineral and mineral name were approved, prior to publication, by the Commission on New Minerals and Mineral Names of the International Mineralogical Association. The distribution of the type material, a part of regolith breccia 14076, is supervised by the lunar sample curator, NASA Johnson Space Center, Houston, Texas.

METHODS

Thin-section studies

Thin section 14076,5, prepared by the lunar-sample curation facility at Johnson Space Center, Houston, was used for these studies. Optical data were obtained using Olympus and Zeiss petrographic microscopes. All U-stage work was done with a Zeiss four-axis stage. Planar features that intersect the *c*-axis were indexed based on the angle of intersection and the calculated cell dimensions.

Electron microprobe analyses were obtained with a Cameca Camebax electron microprobe with a Tracor Northern electronic system. Microprobe automation is described by Chambers (1985); data reduction used the modified Bence-Albee empirical correction factors of Chambers (personal communication). For the data listed in Tables 1 and 2, minimum detection limits (99.7% confidence level) are 0.12% for Na_2O , 0.10% for MgO , 0.12% for TiO_2 , and 0.10% for FeO . Relative errors (1σ) are 0.9% for Al_2O_3 , 0.8% for SiO_2 , and 1.0% for CaO . Relative errors for MgO range from 1.5% at 2% concentration to 8% at 0.2% concentration; for TiO_2 the relative error range is from 11% at 0.2% to 16% at 0.1% concentration; for Na_2O and FeO the relative error range is from 4% at 1.2% concentration to 14% at 0.2% concentration.

An ISI DS-130 scanning electron microscope (SEM) with Tracor Northern energy-dispersive detectors was used to make step-scans across the crystals and the devitrified glasses in order to search for zonation and other compositional variations.

Mineral separations

A 101-mg coarsely powdered split of sample 14076,1 was examined for separable fragments of the new min-

eral. This sample split was first sieved into $>75\ \mu\text{m}$, 75–45 μm , and $<45\ \mu\text{m}$ fractions using ethanol, and the two coarsest fractions were examined in air using petrographic and binocular microscopes. Hand-picked grains or unpicked clusters of grains were mounted on transparent tape or in collodion on aluminum stubs and carbon-coated for energy-dispersive (EDS) chemical analysis using the SEM. The new mineral has a distinctively high Al-Si ratio, and the EDS method was found to be very effective for detecting any anorthite fragments unintentionally included in separates of the new mineral. Following this analysis, a clean concentration of the new mineral was prepared. Forty-seven fragments, each 45 to 150 μm in diameter were selected. These fragments were mounted for X-ray diffraction (XRD) analysis; it was decided not to crush the fragments because they are of more value if left intact for future XRD and transmission electron microscopy (TEM) studies. The sample masses analyzed by XRD were determined using SEM observations of size and the values of density determined by Yoshioka (1970a); these masses were 0.06 mg in the first long-duration powder diffractometer run and 0.08 mg in the second.

X-ray diffraction analysis

Both Guinier camera and powder-diffractometer methods were used to study this small sample. Scans of up to three days with the Guinier camera gave rise only to a few useful diffraction lines. An attempt was made to analyze a single 40- μm crystal using a four-circle diffractometer, but no reflections were detected. Better results were obtained with a Siemens powder diffractometer and Kevex solid-state detector. The sample was mounted on a zero-background quartz plate; in order to get useful peak-to-background ratios, data were obtained in increments of $0.02^\circ\ 2\theta$ with measurement times of six minutes per step. Two powder-diffractometer scans were made; the longest (from 9 to $70^\circ\ 2\theta$) required ten days. Lattice parameters were refined using the XRD data and the program LCLSQ (Burnham, 1965).

PETROGRAPHY

The portion of regolith breccia 14076 that contains yoshiokaite is not representative of the entire regolith breccia. Studies of another powder and another thin section from the other portion of 14076 reveal a different composition and no yoshiokaite. The entire sample is a complex regolith breccia, made up of two very different regolith types. Chemical studies of both parts of sample 14076 are summarized in Jerde et al. (1990), and a description of the glass constituents is given by Vaniman (1990). The present paper treats only that part of the regolith breccia containing yoshiokaite.

The portion of the regolith breccia containing yoshiokaite has no agglutinates (vesicular pieces of glass-bonded soil) and has very few glass spheres (Vaniman et al., 1988; Vaniman, 1990). The few spheres that have been found are sufficient to provide confirmation that the sample is a regolith breccia and not another type of higher-

grade impact breccia. The absence of any agglutinate fragments and the very small I_p/FeO ratio both indicate that this regolith breccia is ancient. McKay et al. (1986) suggested that Apollo 16 regolith breccias with these features were formed about 4×10^9 years ago, during the period of major bombardment of the Moon.

The ultimate origins of yoshiokaite are difficult to trace. The Apollo 14 landing site was purposefully placed on an ejecta ridge that radiates from the Imbrium impact basin. It was hoped that the site would provide samples from deep in the lunar crust. In fact, the ejecta appear to be so well homogenized that extremes in composition are rare. The small sample containing yoshiokaite is among the exceptions.

Yoshiokaite occurs in sample 14076 both as single crystal grains and as small intergrown crystals within devitrified glasses. Most abundant (12 of the 18 particles studied in thin section) are fragments of devitrified glass. These fragments are angular, often fractured, and range up to 190 μm in diameter. Devitrification textures are generally granular. Other glasses with ~ 34 wt% SiO_2 (Table 1b, particles 7, 14, and 54) have plumose to spherulitic devitrification textures (Fig. 1A). Yoshioka (1970a, 1970c) found that the nepheline-like structure formed readily in glasses with silica contents between 9 wt% and 30 wt% SiO_2 ; for SiO_2 contents below 9 wt% a kalsilite-like structure formed (Yoshioka, 1970c) and above 30 wt% an unknown phase formed. In this paper the unknown phase will be referred to as the Ω phase. It is possible that the devitrified lunar glasses with 34 wt% SiO_2 (Fig. 1A) contain the Ω phase; however, the small fragments separated for XRD analysis in this study have average cell parameters that correspond to an average SiO_2 content of 29 wt%, within the compositional range of the nepheline-like structure. No XRD data were obtained for the fragments with 34 wt% SiO_2 and no single crystals were found with more than 28 wt% SiO_2 . The identification of minerals in devitrified glasses with 34 wt% SiO_2 (Fig. 1A; Table 1b, particles 7, 14, and 54) must therefore await further analysis.

The devitrified glasses with 30 wt% SiO_2 or less (Table 1b; Fig. 1B) have compositions and textures similar to those in which Yoshioka (1970a) synthesized nepheline-like structures. Individual crystals in these devitrified glasses are small, angular, and subequant with grain sizes of ~ 5 to 15 μm . In most of the lunar fragments, the crystallite sizes are smaller near the rim of the devitrified glass (Fig. 1B). This gradation of devitrification texture suggests that most of the angular lunar glasses of this type cooled and devitrified in their present form and are not broken remnants of a larger devitrified mass.

Six of the 18 yoshiokaite particles found in thin section are single crystals ranging in size from 45 to 235 μm . These crystals are generally fractured (Fig. 1C) and have patchy to undulatory extinction (Fig. 1D). Sets of lamellae that may be shock-induced occur in most of the crystals; a set of $\{201\}$ lamellae occurs in crystal 63 (Fig. 1E), and well-formed sets of $\{201\}$ and $\{100\}$ lamellae occur

in crystal 52 (Fig. 1F). Lack of variation in c -axis orientation between these lamellae, plus the visibility of the boundaries between them (which would not be expected in twinning where the c -axes remain parallel), suggests that the lamellae are not related by twinning. Multi-element SEM scans across the crystal with the most pervasive lamellar structure (Fig. 1F) reveal slight variations in $Al/(Ca + Si)$ ratios between lamellae; these variations are not seen in crystals without lamellae and may result from shock melting. It is unlikely that the chemical differences associated with the lamellae are due to exsolution because yoshiokaite is metastable and should break down to other phases on prolonged thermal annealing (Yoshioka, 1970a). The postulated shock melting may be similar to that observed in quartz (Gratz et al., 1988), but it occurs as a partial melt rather than as a complete melt that is redistributed along lamellar zones. Indeed, partial melts with large values of $(Ca + Si)/Al$ may be expected on the basis of the liquidus topography around yoshiokaite-like compositions (Fig. 2). Analysis by TEM will be necessary to test this suggestion of association of shock deformation with partial melting.

Crystals larger than 50 μm are severely fractured; the same is true of the devitrified glasses. There is no direct evidence relating these shock effects to the last impact events involved with the formation of regolith breccia 14076, but the degree of fracturing of some of the larger fragments suggests that they would not have remained intact if the fracturing were acquired before they were incorporated into the regolith breccia. The porosity of thin section 14076,5 determined by SEM image analysis is 15%; this is low in comparison with the other part of regolith breccia 14076 (porosity 26%), the part that is like the local Apollo 14 soils. A regolith breccia porosity of 15% is quite small; McKay et al. (1986) found regolith breccias with calculated porosities as low as 17% at the Apollo 16 site, but most calculated and SEM-determined porosities are between 20 and 30%. Low porosity indicates that the yoshiokaite-bearing sample is very compact, suggesting a relatively high degree of shock compaction, thermal sintering, or both. At least some of the shattering and deformation of the yoshiokaite samples apparently occurred during formation of the regolith breccia.

CHEMISTRY

Chemical compositions of yoshiokaite crystals (Table 1a) and of yoshiokaite-bearing devitrified glasses (Table 1b) are very similar. There are, however, some important caveats in the comparison of chemical data for crystals and devitrified glass. Analytical SEM step scans across yoshiokaite crystals detect no chemical variation or zonation, with the exception of crystal 52 in which $Al/(Ca + Si)$ ratios vary between probable shock lamellae. Similar step scans across devitrified glasses indicate that certain elements are excluded from the yoshiokaite grains during devitrification, with the nature of the excluded elements dependent on the silica content of the glass.

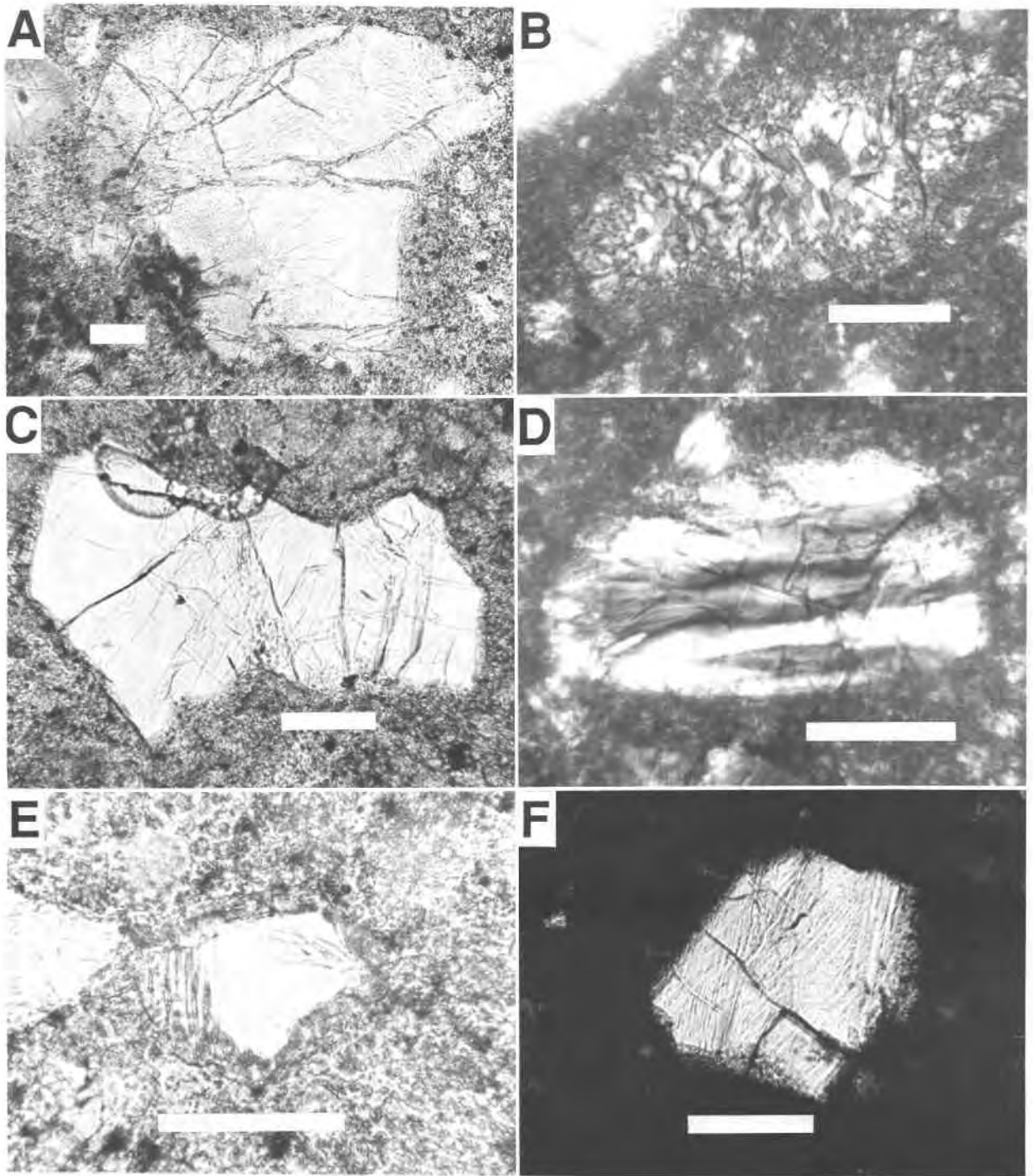


Fig. 1. Photomicrographs. White scale bar in each photomicrograph is 50 μm long. (A) Plumose to spherulitic relatively high-silica glass (34 wt% SiO_2) of uncertain mineral content (particle #14 in Table 1b). (B) Granular devitrified silica-poor (22 wt% SiO_2) glass with anhedral interlocking crystals of yoshiokaite (particle #18 in Table 1b); note gradation from relatively coarse (5–15 μm) grains to finer (1–2 μm) grains at the rim of the devitrified glass (crossed polarizers). (C) Largest yoshiokaite crystal

yet found (grain #23 in Table 1a); note fracturing probably caused by shock deformation. (D) Streaked and patchy birefringence in shocked yoshiokaite crystal (grain #29 in Table 1a; crossed polarizers). (E) shock lamellae of $\{201\}$ orientation in small yoshiokaite crystal (grain #63 in Table 1a). (F) sets of $\{201\}$ and $\{100\}$ shock lamellae in yoshiokaite crystal #52 (Table 1a), crossed by poorly developed $\{100\}$ cleavage (dark fractures trending NW; crossed polarizers).

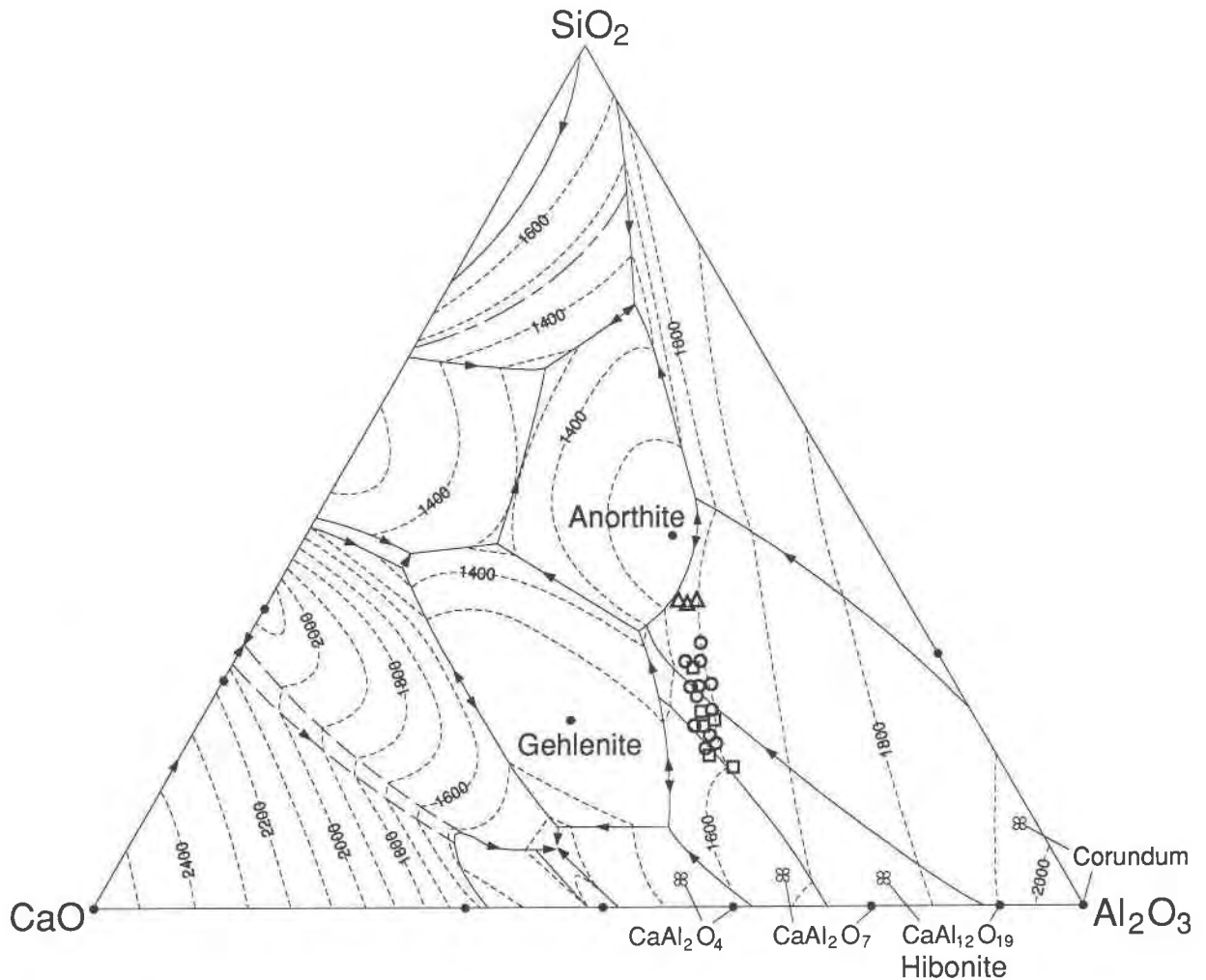


Fig. 2. Ternary diagram showing the compositions of yoshiokaite crystals (open squares) and of devitrified yoshiokaite-bearing glasses (open circles) in the system CaO-Al₂O₃-SiO₂. Triangles mark the compositions of devitrified glasses with ~34 wt% SiO₂ (possible Ω phase). The phase diagram is based on that of Levin et al. (1964) with modifications near the Al₂O₃ apex (Levin et

al., 1969); liquidus temperature contours are in degrees Celsius. All representations are in weight percent. Several Ca,Al-oxide compositions discussed in the text are listed along the lower edge of the figure; lines connect to their bulk compositions (solid dots) and to cloverleaf symbols in corresponding primary liquidus fields.

The relatively high-silica glasses (34 wt% SiO₂; Table 1b) form plumose or spherulitic crystals (Fig. 1A) that are enriched in Al and Si but depleted in Mg, Fe, and Ti relative to the intergranular regions of the devitrified glass. These relatively high-silica devitrified glasses may contain the Ω phase, but their mineral content is still uncertain.

At lower silica contents (30 wt% to 27 wt%), the glasses devitrified to form yoshiokaite grains that are enriched in Ca but have lower Mg, Al, and (to a lesser extent) Si contents relative to the other phases. Devitrified glass with silica content less than 27 wt% also shows evidence of intergranular regions with varied chemistry as observed in backscattered electron images, but such regions are

generally very small (<0.5 μ m), and no significant chemical variation was detected by electron-microbeam methods. Analyses of devitrified yoshiokaite-bearing glasses, as listed in Table 1b, should be considered as analyses of bulk impact melts rather than as analyses of yoshiokaite. Although the chemical differences between the devitrified glass fragments and the single crystal fragments are slight, only the analyses in Table 1a correspond to yoshiokaite.

The yoshiokaite-bearing portion of regolith breccia 14076 is chemically unique compared to other samples from the Apollo 14 site. Most regolith breccias collected from the Apollo 14 site are similar in composition to the Apollo 14 soils. The average Apollo 14 regolith breccia has SiO₂, ~47.6 wt%; TiO₂, ~1.55 wt%; Al₂O₃, ~17.6

TABLE 1A. Yoshiokaite mineral analyses from thin section 14076,5

	Grain no.					
	82	29	52	81	23	63
Size (μm)	45 × 30	115 × 50	95 × 70	70 × 35	235 × 95	50 × 25
SiO ₂	28.0	22.6	21.6	20.6	18.4	15.8
TiO ₂	0.07	0.02	0.08	0.14	0.14	0.12
Al ₂ O ₃	45.9	50.0	52.2	51.4	53.3	56.3
FeO	0.18	0.07	0.00	0.08	0.05	0.41
MgO	0.98	0.27	0.09	0.30	0.22	0.40
CaO	25.5	27.7	26.5	28.3	27.8	27.8
Na ₂ O	0.00	0.02	0.02	0.00	0.00	0.09
K ₂ O	0.00	0.00	0.01	0.00	0.00	0.01
Σ	100.7	100.6	100.6	100.8	99.9	100.9
Cations and vacancies (□) based on 32 oxygens						
Si	5.38	4.42	4.22	4.05	3.64	3.12
Ti	0.01	0.00	0.01	0.02	0.02	0.02
Al	10.42	11.51	11.98	11.88	12.46	13.11
□	2.69	2.21	2.11	2.03	1.82	1.56
Fe	0.03	0.01	0.00	0.01	0.01	0.07
Mg	0.28	0.08	0.03	0.09	0.06	0.12
Ca	5.27	5.80	5.54	5.94	5.92	5.87
Na	0.00	0.01	0.01	0.00	0.00	0.04
K	0.00	0.00	0.00	0.00	0.00	0.00
Σ	24.08	24.04	23.90	24.02	23.93	23.91

wt%; FeO, ~10.5 wt%; MgO, ~9.6 wt%; CaO, ~11.0 wt%; Na₂O, ~0.74 wt%; and K₂O, ~0.56 wt% (average of 7 regolith breccias analyzed by Fruland, 1983); within analytical uncertainty, this is identical to the composition of representative Apollo 14 soil 14163 (Papike et al., 1982). This regolith composition has more Si, Ti, Fe, Mg, Na, and K and less Ca and Al than the yoshiokaite-bearing sample 14076,1 (Warren et al., 1987; Jerde et al., 1990). The bulk chemistry of the yoshiokaite-bearing sample reflects both the contribution of yoshiokaite to the sample and the absence of typical Apollo 14 soil (Fra Mauro). This composition is represented in other Apollo 14 regolith breccias and in Apollo 14 soils by a variety of impact glass with composition similar to the bulk Fra Mauro soil. It is both the presence of yoshiokaite and the lack of typical Fra Mauro glass fragments (Vaniman, 1990) that makes the yoshiokaite-bearing part of regolith breccia 14076 chemically unique.

It would be difficult to create the very low silica content of both yoshiokaite crystals (Table 1a) and yoshiokaite-bearing devitrified glasses (Table 1b) by any process other than Si loss through volatilization during heating caused by impact. All of the compositions in Table 1 can be represented in the ternary system CaO-Al₂O₃-SiO₂ (Fig. 2); not one of the crystals or devitrified glasses referred to in Tables 1a and 1b has more than 3 wt% of other chemical components. Yoshiokaite crystal and glass compositions plotted in this system lie below anorthite, along a trend of silica loss. This trend crosses the primary liquidus fields of corundum and hibonite, almost parallel to the liquidus isotherms and crossing at least one reaction curve (between corundum and hibonite) at a high angle. Such a trend can not be generated by equilibrium or fractional crystallization processes.

The alignment of the yoshiokaite trend with anorthite and silica in Figure 2 suggests Si loss from a system with

an anorthositic composition as a possible origin. Fractional vaporization of K₂O > Na₂O > SiO₂ > Fe has been widely accepted as an explanation of much of the variation in composition of lunar impact glasses (Naney et al., 1976; Delano et al., 1981). Loss of silica by vaporization was implicated in the formation of HASP glasses by Naney et al. (1976). Although this effect can be seen in many impact glasses, the most prominent examples are glasses from the lunar highlands that have high alumina (the weight-percent ratio of CaO/Al₂O₃ is ~0.5–0.6) and low silica (SiO₂ is <40 wt%). Table 2 summarizes the available data on these HASP-like glasses. Comparison of the data in Table 1 with those in Table 2 shows that the yoshiokaite samples are generally higher in Ca and Al at a given Si content than the other HASP-like samples; the yoshiokaite samples also have less Mg and Fe. The only HASP-like glasses similar to the yoshiokaite samples are either from the same lunar landing site (Apollo 14 HASP glass F5, Table 2a) or from other, possibly ancient regolith breccias (average HASP from Apollo 16 regolith breccias, Table 2b).

One problem with the impact-volatilization hypothesis is the occurrence of measurable sodium (up to 0.4 wt% Na₂O), particularly in the yoshiokaite-bearing devitrified glasses with 25–28 wt% SiO₂. No other HASP-like compositions contain so much sodium, and sodium should be lost much more readily than silica where impact-volatilization occurs in vacuum. Na may be retained during vaporization of Si if the impact vaporization occurs within a highly oxidizing gas (Walter, 1967), but such a situation is unlikely to occur on the Moon. Alternatively, the source rock for these particular yoshiokaite-bearing devitrified impact glasses may have been exceptionally Na-rich. There is a strong indication, as discussed below, that impact into anorthositic crust is required to form the glasses described here. The most abundant anorthosite

TABLE 1B. Silica-poor polycrystalline devitrified glasses from sample 14076,5

	Particle no.								
	Ω phase (?)			yoshiokaite-bearing					
	7	54	14	51	43	13	32	80	60
Size (μm)	50 × 45	60 × 50	340 × 310	160 × 70	70 × 50	120 × 120	45 × 35	70 × 35	90 × 65
SiO ₂	34.4	34.3	34.1	30.0	28.0	27.4	25.5	25.2	24.8
TiO ₂	0.12	0.08	0.23	0.16	0.09	0.08	0.08	0.10	0.07
Al ₂ O ₃	42.7	40.6	41.0	45.8	47.0	46.4	49.2	48.1	48.7
FeO	0.66	1.21	0.33	0.00	0.00	0.04	0.04	0.07	0.06
MgO	0.57	1.87	2.10	0.69	0.18	0.59	0.76	0.26	0.20
CaO	21.1	22.8	22.2	23.5	24.9	24.9	24.9	27.0	26.9
Na ₂ O	0.01	0.02	0.02	0.13	0.40	0.16	0.30	0.17	0.06
K ₂ O	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.01
Σ	99.6	100.9	100.0	100.3	100.6	99.6	100.8	100.9	100.8

fragments at the Apollo 14 site are alkali anorthosites with ~1.5–2.5 wt% Na₂O; impact volatilization could cause removal of most of the Na from such an alkali-rich target material with detectable Na still left in the impact melt.

PHYSICAL AND OPTICAL PROPERTIES

Although yoshiokaite is hexagonal (Yoshioka, 1970a), most of the crystals in regolith breccia 14076 have anomalous optical properties (small $2V$, $<5^\circ$) due to strain. All grains are optically positive. Birefringence is ~0.02 or less, and single crystals generally have undulatory or patchy extinction (Fig. 1D). Because the optical properties of the natural samples have been affected by strain, we relied on the optical data of Yoshioka (1970a) in compiling Figure 3. Yoshioka (1970a) reported no characteristic twinning or cleavage in his synthetic samples; among the natural samples there is one crystal with poorly developed {100} cleavage (Fig. 1F). This crystal is also the one with most extensive shock lamellae.

In the devitrified glasses with 30 wt% SiO₂ or less (Fig. 1B), crystals of yoshiokaite are interlocking, angular, and anhedral, with no preferred orientation (Fig. 1B). The devitrified glasses with 34 wt% SiO₂ (Fig. 1A) have spherulites of length-slow (positive elongation) fibers and plumose crystals with inclined extinction; these spherulitic

TABLE 2A. High Al,Si-poor glass compositions in other Apollo 14 samples

	Sample								
	14259*	F2**	F3**	F4**	F5**	F6**	F7**	F8**	F9**
SiO ₂	33.7	31	30	37	29	32	29	36	36
TiO ₂	0.24	0	0	0	0.5	0.5	0	0	0
Al ₂ O ₃	35.2	41	46	37	47	37	40	34	39
FeO	0.72	0	0	0	0	0.5	11	1	0
MgO	8.04	3	6	1	0	8	2	8	8
CaO	20.7	24	19	23	26	22	12	19	24
Na ₂ O	0.01	0	0.5	0	0	0	0	0	0.5
K ₂ O	0.00	0	0	0	0	0	0	0	0
Σ	98.6	99	101.5	98	102.5	100	94	98	107.5

* Analysis by Brown et al., 1971; ** Analyses by Finkelman, 1973 (semi-quantitative).

and plumose crystals may consist of the Ω phase, but the minerals have not been identified yet.

Values of density for synthetic yoshiokaite plotted in Figure 3 are also taken from the work of Yoshioka (1970a). Our sample of 0.08 mg collected from 14076,1 contains few single crystals. These crystals are too small for accurate density determinations and may have anomalous values of density owing to shock deformation in any event. The data in Figure 3 indicate that the density decreases significantly as silica content (and hence vacancy content) increases. We have added to Figure 3 the estimated viscosity of yoshiokaite liquids at 1600 °C, which is slightly greater than the liquidus temperature (Fig. 2). Viscosities are quite small and may well affect processes such as melt-sheet flowage and thinning in impact melts of yoshiokaite composition.

CRYSTALLOGRAPHY

X-ray diffraction data confirm that Yoshioka's synthetic phase has been found in nature. Figure 4 shows two

TABLE 2B. High Al,Si-poor glass compositions from other lunar areas

	Sample						
	Apollo 11*		Apollo 16			Apollo 17	Luna 24
	soil 10019	soil 10094	soil av.**	soil av.†	r.b. av.‡	soil av.§	soil av.
SiO ₂	35.1	33.5	30.8	31.9	28.7	30.9	38.5
TiO ₂	0.23	0.18	0.67	0.82	0.16	1.44	0.31
Al ₂ O ₃	34.0	34.9	38.0	34.2	43.9	31.2	30.4
FeO	0.34	0.43	2.84	4.98	0.37	6.01	8.81
MgO	8.56	10.92	6.02	8.49	2.5	12.0	6.98
CaO	20.2	20.7	21.6	19.5	24.4	18.7	17.1
Na ₂ O	0.00	0.02	0.03	0.13	0.00	0.00	0.01
K ₂ O	0.01	0.00	0.02	n.a.	0.01	0.00	n.a.
Σ	98.4	100.6	100.0	100.0	100.0	100.2	102.1

Note: r.b. = regolith breccia (all other samples are from soils); n.a. = not analyzed.

* Analyses by Simon et al., 1984.

** Average of 12 small glass beads (10–20 μm) analyzed by Kempa and Papike, 1980.

† Average of five glass beads analyzed by Naney et al., 1976.

‡ Average of three glass beads in regolith breccias analyzed by Heavilon and Basu, 1988.

§ Average of three glass beads analyzed by Vaniman and Papike, 1977.

|| Average of three glass beads analyzed by Norman et al., 1978.

TABLE 1B—Continued

Particle no.					
yoshiokaite-bearing					
65	18	41	21	33	64
25 × 25	190 × 85	90 × 55	165 × 150	90 × 50	35 × 35
23.9	22.3	20.5	19.3	18.6	17.5
0.12	0.09	0.11	0.09	0.08	0.13
48.8	51.2	51.1	52.4	53.5	53.8
0.13	0.08	0.02	0.02	0.00	0.11
0.53	0.37	0.15	0.26	0.09	0.24
26.6	26.2	28.9	27.8	28.5	29.1
0.04	0.04	0.01	0.00	0.01	0.03
0.02	0.00	0.00	0.00	0.00	0.00
100.1	100.3	100.8	99.9	100.8	100.9

XRD patterns obtained from yoshiokaite mineral grains and devitrified glasses that were hand-picked from sample 14076,1. Both patterns were obtained using a zero-background quartz plate. The adhesive-tape mounting medium is responsible for the high background intensity in the range $2\theta = 13$ to 26° in the upper diffraction pattern. Use of light machine oil as a mounting medium resulted in lower background intensities. Comparison of the two diffraction patterns (Fig. 4, Table 3) reveals large differences in relative peak intensities, largely owing to orientation variations in the small number of unpowdered fragments that were used.

The greatest accumulated mass of yoshiokaite weighs only 0.08 mg; despite this limitation, the similarity in XRD patterns of natural yoshiokaite and Yoshioka's synthetic crystals (1970a), when considered with the chemical data, demonstrates that synthetic and natural phases are equivalent. Some of the discrepancies between our data and those of Yoshioka are no doubt related to (1) sample orientation effects in our unpowdered fragments, (2) the small mass of sample used in this study, and (3) the fact that our combined concentrate covers a large range in solid solution (probably 30 wt% to 16 wt% SiO_2 ; Tables 1a and 1b) whereas data of Yoshioka are for a specific composition with ~ 20 wt% SiO_2 . The small fragments analyzed in this study by XRD may well have contained some of the Ω phase; if so, then there was insufficient Ω phase to be detected, or it has diffraction characteristics similar to those of yoshiokaite.

Yoshioka (1970a) defined this new phase as a nepheline-type solid solution with composition on the CaAl_2O_4 -anorthite join (Fig. 2). Later synthesis and study of the single composition $\text{CaAl}_2\text{SiO}_6$, which is actually within Yoshioka's solid solution series, suggested that it has space group $P3c1$ or $P\bar{3}c1$ (Kirkpatrick and Steele, 1973). Recent single-crystal analysis of this synthetic material by Steele and Pluth (1989) showed that it has a very distorted stuffed-tridymite structure; their work in progress suggests that the correct space group corresponding to this synthetic composition, and by inference yoshiokaite, is $P\bar{3}$ (I. M. Steele, personal communication, 1989). Calculated diffraction intensities, using cell parameters of this study and the structure of Steele and Pluth with space group $P\bar{3}$

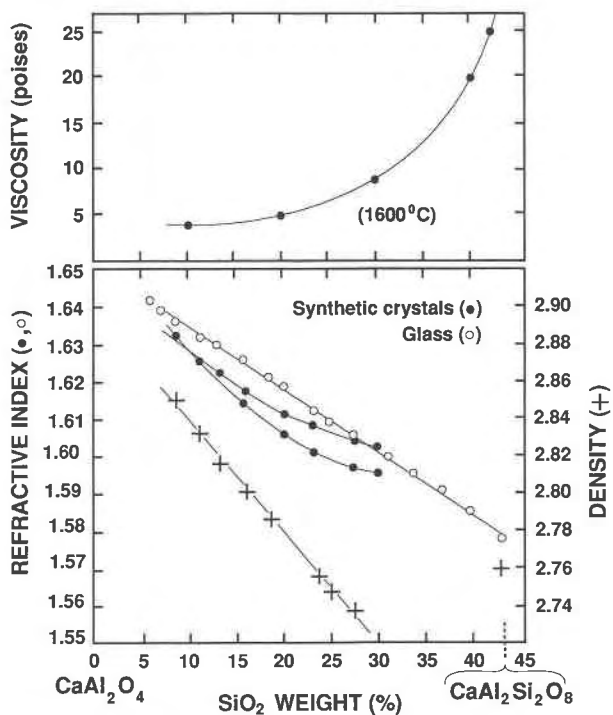


Fig. 3. Refractive index and density data for unshocked synthetic equivalents of yoshiokaite and for anorthite (after Yoshioka, 1970a). Melt viscosity data are for equivalent liquid compositions at 1600°C (Ryan and Blevins, 1987).

(Table 3), are very similar to those of the synthetic sample (Yoshioka, 1970a). The Miller indices assigned in Table 3 differ in some cases from those of Yoshioka (1970a).

Yoshioka's formula for the solid-solution series can be written as $\text{Ca}_{8-(x/2)}\square_{(x/2)}\text{Al}_{16-x}\text{Si}_x\text{O}_{32}$, with Ca filling some of the interstitial sites in a stuffed derivative of the high tridymite structure. Yoshioka (1970c) suggested that the nepheline-like structure will form if $x = 2$ –5.3. For $x < 2$, a kalsilite-like structure forms, and for $x > 5.3$, the Ω phase forms. Among the lunar yoshiokaite fragments, only the six single crystals analyzed in thin section permit a test of this range in x . For those analyses, values of x range from 3.1 to 5.4 (Table 1a). The actual range of x in lunar yoshiokaite is uncertain owing to the limited number of crystal fragments and the uncertain mineral content of devitrified glasses with 34 wt% SiO_2 (Table 1b).

The solid solution in yoshiokaite involves a coupled exchange between CaAl_2 and $\square\text{Si}_2$. Thus, a major difference between ordinary nepheline-like structures and that of yoshiokaite involves the large variation in Si-Al ratio and the potentially large number of vacant sites at larger Si:(Ca + Al) ratios. Presumably, Ca is accommodated in the Na sites of nepheline [$\text{Na}_3\text{K}(\text{Si}_4\text{Al}_4\text{O}_{16})$] with the K sites being vacant. The maximum Ca:(Si + Al) ratio should thus be 3:8, which is lower than the ratio 1:2 of the Si-absent end-member CaAl_2O_4 . The smallest SiO_2

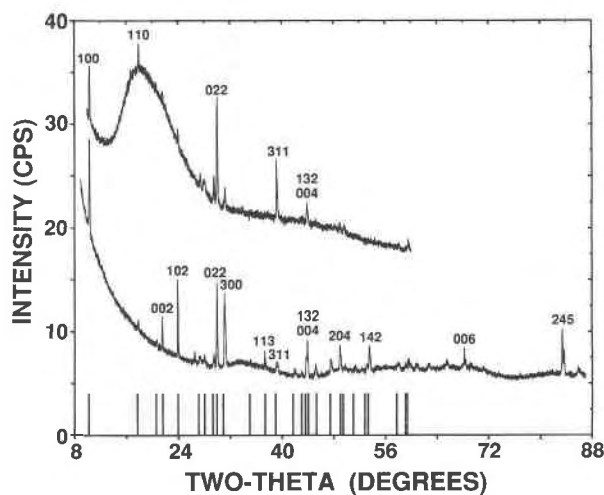


Fig. 4. X-ray diffraction patterns of yoshiokaite crystals and yoshiokaite-bearing devitrified glasses concentrated from sample 14076.1, compared with the diffraction lines (vertical lines above 2θ axis) obtained by Yoshioka (1970a). The upper pattern was obtained with a 0.06-mg sample held to the zero-background quartz plate by a small patch of transparent adhesive tape (note elevated background from about 13 to 26° 2θ); the lower pattern was obtained with a 0.08-mg sample in a minimal amount of light machine oil (only slightly elevated background from about 32 to 38° 2θ). Labeled peaks illustrate variable intensities owing to differences in sample orientation between the two scans. The upper diffraction pattern is shifted upward to separate the two patterns.

content yet observed for lunar yoshiokaite is about 16 wt%, which corresponds to $\text{Ca}:(\text{Si} + \text{Al}) \approx 3:8$ (Table 1a). Yoshioka (1970a) was able to synthesize crystals of this structure with SiO_2 content as small as 9 wt% and $\text{Ca}:(\text{Si} + \text{Al})$ as large as 7:16. However, he also noticed a sharp discontinuity in the variation of a and c as a function of composition, where the ratio $\text{Ca}:(\text{Si} + \text{Al}) = 3:8$ is exceeded. Based on Yoshioka's experiments, there is reason to believe that lunar yoshiokaite cannot have less than ~16 wt% SiO_2 .

Yoshioka (1970a) determined that the c cell parameter of his synthetic crystals was smaller (~8.2 Å) than that of nepheline (~8.4 Å). In a later study of systems in which Na and K were added, Yoshioka (1970b) found that the synthetic structure with the smaller value of c persisted only for compositions for which the $(\text{Na,K})\text{AlSiO}_4$ content was less than ~10 wt%. The lunar yoshiokaite samples have little alkali, such that the composition of end-member nepheline is not approached in these natural samples.

DISCUSSION: THE PROBABLE ORIGINS OF LUNAR YOSHIOKAITE

Synthetic crystals identical to yoshiokaite are formed by melting an appropriate composition, quenching the melt to form a glass, and then crushing the glass and maintaining it at a low subsolidus temperature from sev-

TABLE 3. XRD data for yoshiokaite, the synthetic phase (Yoshioka, 1970a), and calculated intensities for a structure with space group $P\bar{3}$

hkl	Scan 1 yoshiokaite*		Scan 2 yoshiokaite**		Yoshioka (1970a)		Calcu- lated intensi- ty†	
	d(obs)	I(obs)	d(calc)	I(obs)	d(obs)	I(obs)		
100	8.54	50	8.61	8.57	100	8.68	31	51
110	4.94	26	4.97	4.97	10	5.00	36	43
200	4.29	9	4.30	4.30	7	4.31	2	4
002	4.105	12	4.122	4.123	36	4.11	22	24
102	3.709	15	3.718	3.718	79	3.71	9	8
210	3.251	14	3.253	3.254	9	3.280	28	25
112	3.168	13	3.173	3.176	10	3.175	11	10
121, 211	3.019	24	3.026	3.026	13	3.041	55	53
022	2.972	100	2.977	2.979	91	2.980	100	100
300	2.862	20	2.869	2.871	78	2.886	39	32
212			2.554	2.554	4	2.560	5	4
113, 310			2.405	2.405	16	2.402	8	6
311	2.297	54	2.293	2.295	11	2.306	35	29
400			2.152	2.153	8	2.165	4	3
213	2.093	4	2.099	2.101	4	2.101	15	12
132			2.066	2.069	20	2.074	26	26
004	2.061	20	2.061	2.062	41	2.055	12	25
014	2.000	4	2.005	2.005	13	2.000	11	9
402			1.908	1.909	15	1.916	3	3
204	1.857	8	1.859	1.860	32	1.856	18	16
411	1.830	8	1.831	1.830	3	1.841	11	8
322			1.781	1.777	6	1.788	10	8
500			1.722	1.721	6	1.731	3	2
142			1.709	1.710	28	1.716	41	6
052	1.584	3	1.589	1.588	6	1.595	13	11
314			1.560	1.561	5	1.561	6	5
413, 510						1.555	15	
413	1.551	10	1.551	1.551	10			9
510			1.546	1.547	7			8
151			1.520	1.520	7			6
234			1.426	1.426	4			4
006, 520			1.374	1.373	20			24
135			1.357	1.357	3			9
504			1.321	1.322	3			6
252, 206			1.307	1.308	3			4
604, 702			1.178	1.177	2			8
245, 406			1.158	1.158	51			3
443			1.132	1.131	9			4

Note: Calculated cell parameters for the lunar mineral using scan 2: $a = 9.939(2)$ Å, $c = 8.245(2)$ Å, $V = 705.4(3)$ Å³.

* Sample mounted with plastic tape.

** Sample mounted with light machine oil.

† Based on a structure with space group $P\bar{3}$ (Steele and Pluth, 1989) and cell parameters of this study.

eral minutes to several days (Yoshioka, 1970a). The temperature at which the nepheline structure-type phase appears ranges from 950 to 1200 °C. This phase decomposes if it is treated at higher subsolidus temperatures or if it is held at its temperature of appearance for more than a few days or weeks.

The melting and cooling conditions for this mode of formation are readily produced by impact melting. In an impact-melt sheet, the initial superheat is quickly lost to cold clasts, and the melt rapidly (within tens of seconds) reaches an equilibrium temperature of about 1200–1600 °C (Simonds et al., 1976). At this point a period of second-stage cooling begins, which can range from minutes to days or longer depending on the mass of the glass fragment or the thickness of a glassy melt sheet. However,

thick impact-melt sheets may cool too slowly to permit formation of metastable minerals such as yoshiokaite.

Studies by Onorato et al. (1976) seem to rule out the formation of yoshiokaite in thick melt sheets. Approximately 36 d are required for cooling of the top of a 10-m-thick melt sheet, covered by 1 m of cold debris; a year is required for cooling of the deeper zones to below 900 °C. These cooling rates are probably too slow to allow for preservation of metastable yoshiokaite crystals. The most favorable lunar conditions for formation of yoshiokaite correspond to the top of an uninsulated melt sheet (which should cool to subsolidus temperatures in 2 to 3 d; Onorato et al., 1976) or in small impact-melt coatings, veins, or pods that may cool quickly but without quenching. Formation of at least some yoshiokaite in such small melt bodies is indicated by the decrease in yoshiokaite grain sizes toward the edges of some devitrified glass fragments (Fig. 1B). Small bodies of impact glass may in fact be a prerequisite for yoshiokaite formation on the Moon, since a high surface-to-mass ratio may be needed for extreme Si volatilization. However, the very low viscosity of melts with yoshiokaite-like compositions (Fig. 3) can also give rise to much thinner melt sheets with more rapid cooling rates. Within such melt sheets, yoshiokaite crystallization might be favored even though stable phases (e.g., corundum, gehlenite, and hibonite) might form in thicker portions. This conjecture relies on the assumption that Si-volatilization can occur before or during the emplacement of such melt sheets. Such an assumption is difficult to test, but it is worth noting that crystallization of a Si-depleted anorthositic melt sheet with a range of cooling pathways could link the occurrence of yoshiokaite with its otherwise rare stable equivalents—corundum and possible gehlenite—reported as particles in the soil at the Apollo 14 site.

Formation of yoshiokaite by impact melting is further supported by the chemical variation of Figure 2 and Table 1. Violation of liquidus isotherm and reaction-curve control by this trend rules out equilibrium crystal-liquid control in the origin of the trend. Silica volatilization of an anorthite-rich composition is the more plausible cause. This conclusion is strengthened by the similarity to some other HASP-like compositions that are found in Apollo 14 glasses and in the glasses of Apollo 16 regolith breccias (Table 2, and Vaniman, 1990).

SIGNIFICANCE OF YOSHIOKAITE

Yoshiokaite is probably not a lunar oddity

Other minerals first found on the Moon (armalcolite and tranquillityite) have origins generally coupled to the crystallization of high-Ti basalts; yoshiokaite is the first new mineral with an origin specifically related to the lunar highlands. The very high Ca and Al content of the yoshiokaite-bearing devitrified glasses, combined with their very low Fe and Mg contents, is a strong indication that the target material for making these impact melts was almost pure anorthosite. The retention of measurable

Na in yoshiokaite samples suggests that alkali anorthosites may have been present in the target area.

The anorthositic parentage of yoshiokaite adds to petrologic and remote-sensing evidence that areas of almost pure anorthositic crust are common on the Moon (Davis and Spudis, 1987; Jerde et al., 1990). Unfortunately, the purest anorthositic crust is found mostly on the far side of the Moon (Davis and Spudis, 1987); none of the Apollo or Luna missions directly sampled these areas. The sampled sites are clustered within less than 5% of the lunar surface (Jerde et al., 1990) and are all on the near side. We suggest that even though it was not recognized during the first two decades of lunar sample analysis, yoshiokaite is probably not a rare lunar mineral. That yoshiokaite was not recognized until now is likely not because of its rarity but rather because of our limited sampling of the Moon. Future sampling of the anorthositic limbs and far side of the Moon may well show that yoshiokaite has a widespread distribution.

New minerals to be found on the Moon?

It does not require much speculation to propose that the discovery of yoshiokaite points to other phases that may occur on the Moon. There is the possibility that the Ω phase may be found on the Moon as a new mineral; further studies of the fragments with 34 wt% SiO₂ occurring in the yoshiokaite-bearing portion of regolith breccia 14076 will permit a test of this possibility. There is also no a priori reason to assume that impact melts formed in the anorthositic lunar highlands cannot attain extremely low Si contents (<9 wt% SiO₂) through volatile loss. In such melts a metastable mineral with Yoshioka's (1970c) kalsilite-type structure may form. Furthermore, known minerals that have not yet been reported as occurring on the Moon may be found as stable phases in Si-depleted anorthositic impact melts (e.g., hibonite, Fig. 2).

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