Jadeite and its relatives: fake news or a real tool to constrain shock conditions?

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Abstract

Jadeite is frequently reported in shocked meteorites, displaying a variety of textures and grain sizes that suggest formation by either solid-state transformation or by crystallization from a melt. Sometimes, jadeite has been identified solely on the basis of Raman spectra. Here we argue that additional characterization is needed to confidently identify jadeite and distinguish it from related species. Based on chemical and spectral analysis of three new occurrences, complemented by first-principles calculations, we show that related pyroxenes in the chemical space \((\text{Na})^{M_2}(\text{Al})^{M_1}(\text{Si}_2)^{T_0}_6 - (\text{Ca})^{M_2}(\text{Al})^{M_1}(\text{AlSi})^{T_0}_6 - \square\)^{M_2}(\text{Si})^{M_1}(\text{Si}_2)^{T_0}_6\) with up to 2.25 atoms Si per formula unit have spectral features similar to jadeite. However, their distinct stability fields and synthesis pathways, considered together with textural constraints, have specific implications for precursor phases and estimates of impactor size, encounter velocity, and crater diameter. A reassessment of reported jadeite occurrences puts in a new light many previous conclusions about the shock histories preserved in particular meteorites.

Introduction

Plagioclase is common in Earth’s crust and in many differentiated (e.g., shergottites and eucrites) and undifferentiated meteorites. Under the extreme temperature \((T)\) and pressure \((P)\) conditions that are reached during collisions on and among meteorite parent bodies, sodic plagioclase may transform into a variety of high-pressure (HP) forms, including hollandite-structured lingunitetr13, maskelynite, dialectic glass, jadeite plus a silica phase, or the recently-discovered albitic jadeite45. In any closed-system transformation, albite that decomposes to stoichiometric jadeite must also form excess SiO\(_2\) in the form of stishovite, coesite or an amorphous phase6–9. The phase of SiO\(_2\) that forms depends on peak pressure as well as kinetic factors9–11. Another transformation path, however, is from albite to albitic jadeite—a super-silicic, vacancy-rich pyroxene with excess Si occupying octahedral M1 sites (ref. 4). Tissintite, the Ca-rich analogue of albitic jadeite, forms only from more calcic plagioclase12; it has been found so far only in eucrites and Martian meteorites13,14.

The presence of true stoichiometric jadeite in a given meteorite suggests that it experienced a well-defined set of \(P\) and \(T\) conditions, especially when found in combination with particular high-pressure silica phases. In other contexts, meteoritic jadeite may occur without accompanying silica, suggesting either subsolidus recrystallization or growth from a melt. On the other hand, albitic jadeite and other minerals with related structure have similar physical properties, but unknown and presumably different stability fields. It is therefore critical to ensure that a meteorite contains true jadeite before using its occurrence to constrain shock conditions. It is also an important research goal to experimentally calibrate the shock conditions for the synthesis of albitic jadeite and other forms.

However, distinguishing among jadeite and its relatives is challenging. Several studies have claimed the presence of jadeite on the basis of optical petrography or Raman spectroscopy alone, without confirmation by chemical analysis or a structure-sensitive method such as electron backscatter diffraction (EBSD). One challenge is that the disorder and strain of the crystal due to substitutions and vacancies on certain cation sites can influence the stability of the crystal under irradiation by lasers, X-rays, or high-energy electron beams. Albitic jadeite, for example, is observed to be acutely sensitive to electron beams, becoming amorphous under a focused beam within a few seconds in the scanning electron microscope (SEM) (refs. 4,5) whereas ideal jadeite is stable and retains its structure during microanalysis with focused electron beams. A compositional analysis is clearly required to confirm the Raman identification of jadeite before the stability field of jadeite can be used to set a minimum pressure for a meteorite shock event. However, what are the compositional limits of the recognized jadeite-like species, and how vacancy-rich does a pyroxene have to be to qualify as albitic jadeite or tissintite? We introduce a new ternary diagram for pyroxenes in the space \((\text{Na})^{M_2}(\text{Al})^{M_1}(\text{Si})^{T_0}_6 - (\text{Ca})^{M_2}(\text{Al})^{M_1}(\text{AlSi})^{T_0}_6 - \square\)^{M_2}(\text{Si})^{M_1}(\text{Si}_2)^{T_0}_6\) as a tool to represent the compositional ranges observed. We present new combined textural, chemical, and structural data on jadeite, albitic jadeite, and related phases in shock veins from three L6 ordinary chondrites (Ozerki, Chug Chug 011, and Chantonnay). We then consider these observations in the context of (1) all the published references in which the presence of jadeite and its relatives were reported in chondritic meteorites and (2) new calculations of the Raman spectra of several jadeite-related high-pressure pyroxene compositions. We do not consider tissintite in this work, since it has been so far only observed in achondrites13,14. We suggest that a number of published works based on jadeite occurrences have overestimated the inferred \(P-T\) conditions for shocks on the L-chondrite parent body.
Results

Petrography and mineral chemistry. Ozerki. The studied thin section of Ozerki (see Methods section) displays two discrete areas (Fig. S1A): a light-colored chondritic lithology and a dark-colored lithology dominated by impact melt. We focused on a dense network of dark shock veins that cross-cut the light-colored lithology. The veins are variable in thickness (from 40 to 850 μm) and are locally disrupted by the presence of clasts (up to 1 mm) with angular to sub-rounded shapes. Clasts are more abundant in thicker veins and often display jigsaw-fit breccia textures. The centers of the veins contain mainly silicate clasts, whereas the margins are more rich in globular metal segregations and sulfide grains. Electron microprobe analysis (EPMA) in Ozerki yields a formula for the pyroxene in the range (Na0.49-0.64Ca0.07-0.08)(K0.03-0.05Mg0.00-0.08Fe0.00-0.02)[O0.25-0.40](Al0.77-0.81Si1.14-0.23Fe0.06-0.13)Si2O6, which place this pyroxene close to the albitic jadeite end-member. The Ca# [100×Ca/(Ca+Na)] ranges from 11.0 to 17.6 (Fig. S2). The albitic jadeite is found as acicular to dendritic aggregates of crystallites (up to ~4.5 μm in size) within amorphized plagioclase glass or along plagioclase-pyroxene contacts (Fig. 1). When in contact with matrix pyroxene, the crystallites display core domains with slightly higher electron backscatter brightness, grading into albitic jadeite rims with lower backscatter contrast.

Chug Chug 011. Numerous melt veins crosscut the matrix of the studied section of Chug Chug 011 (Fig. S1B). The veins are about 100 μm wide, with central domains dominated by elongated silicate clasts oriented parallel to the margins of the veins. The pyroxene is found within a zoned clast, featuring a rim of low Ca-pyroxene surrounding sodic plagioclase (Fig. 2). Analysis by energy-dispersive x-ray spectroscopy (EDS) yields an empirical formula (Na0.49-0.64Ca0.07-0.08)(K0.03-0.05Mg0.01-0.024)[O0.25-0.33](Al0.62-0.68Si0.04-0.18Fe0.13-0.21Mg0.21)Si2O6, again close to the albitic jadeite end-member. The Ca# ranges from 10 to 24. As in the Ozerki case, albitic jadeite crystals (~1 μm long and ~200 nm wide) form a rim that completely surrounds plagioclase glass (Fig. 2B).

Chantonnay. Three types of shock veins occur in the studied section of Chantonnay (Fig. S1C): dark-colored clast-rich silicate veins, cross-cut by lighter-colored clast-poor silicate veins, and metal-troilite veins that cross-cut both the silicate veins and the wall rock. More details on the chemistry and textural relationships of matrix minerals can be found in Dodd et al.16. Albitic jadeite is observed within a thick (~300 μm wide) melt vein (Figs. 2C, D). It is co-located with a glassy pool and surrounded by melt vein matrix. Electron microprobe analysis yields an empirical formula (Na0.42Ca0.05Mg0.11)[O0.42](Al0.93Si0.11)Si2O6, with Ca# of 11, whereas EDS analyses suggest a range for Ca# from 19 to 24 (Fig. S2).

The new compositions from all three meteorites are plotted along with available data from the literature on our new ternary diagram for jadeite-related pyroxenes in Fig. 3. The locations of notable molecules and end-members that plot in this ternary diagram include albite [(Na0.75□0.25)M2(A10.75Si0.25)M1Si2O6], anorthite [(Ca0.75□0.25)M2AlM1(Al0.5Si1.5)O6], Ca-Eskola component [(Ca0.5□0.5)M2AlSi2O6] and the hypothetical molecule □M2SiM1Si2O6 (described as "supersilicic pyroxene" by Smith and here called "silipyx"). On this diagram, most clinopyroxenes from the literature and from this study plot inside the Jd–Ab–Ca-Esk triangle. A few analyses plot near the Jd apex, indicating nearly pure jadeite compositions. Most reported compositions, including many assigned to jadeite on the basis of their Raman spectra, clearly display some excess Si, lying between the 2.0 and 2.25 Si atoms per formula unit (apfu) isopleths. It is possible that some of these analyses are contaminated with Si-rich matrix material due to small grain sizes or that beam-sensitive materials have lost Na, but most of the pyroxene crystals in question probably deviate significantly from the Jd–Ca–Esk join. There is no separation on this diagram among ordinary chondrite samples from different groups (LL, L, H).

The data on albitic jadeite from the current study, excluding transmission electron microscope (TEM) data from beam-sensitive material in Chantonnay, display consistent values of excess silica. All the Ozerki analyses but one have Si > 2.125 apfu, and lie close to the Si=2.25 apfu line. The Chug Chug 011 analyses plot closer to the Si=2.125 apfu contour. Along a constant Si contour in this diagram, the fraction of vacant M2 sites increases systematically with increasing Ca content. Ozerki analyses have between 0.25 and 0.40 vacancies per formula unit, whereas Chug Chug 011 analyses range between 0.09 and 0.31 vacancies. All these analyses are consistent with the definition of albitic jadeite as a solid solution with >0.10 vacancies and >2.15 Si apfu.

Raman spectroscopy. Ozerki. The Raman spectra of albitic jadeite in Ozerki display five distinct peaks at 376, 526, 698, 986, and 1036 cm⁻¹ (Figs. 4A, 5). The predominant peak is remarkably consistent from point to point at 698 cm⁻¹. The two peaks at higher wavenumbers related with the vibration of the [Si2O6]4⁻ groups, are visible but not as distinct or well-separated as in the ideal jadeite Raman spectrum (Figs. 4A, 5).
Chug Chug 011. We acquired numerous spectra from the area hosting albitic jadeite crystals in Chug Chug 011. The predominant peak is near 698 cm$^{-1}$ but is shifted to lower wavenumbers (~693 cm$^{-1}$) in some spectra. Moreover, the typical jadeite peak at 1038 cm$^{-1}$ is shifted to 1016 cm$^{-1}$, which may be associated either with a diopside-related structure or another HP clinopyroxene (Figs. 4A, 5). The low-wavenumber peak typically seen at 376 cm$^{-1}$, is shifted to somewhat higher wavenumbers (~388 cm$^{-1}$).

Chantonnay. The Raman spectrum of albitic jadeite in Chantonnay displays three major peaks at 377, 699, and 1038 cm$^{-1}$ alongside less intense peaks at 222, 254, 328, 522, and 987 cm$^{-1}$ (Fig. 4A). The peak positions closely match the reference spectrum of jadeite (RRUFF ID_R050220) in the RRUFF database$^{18}$, but many of them have modestly broader line shapes. This broadening may be associated either with crystals that are small compared to the Raman spot diameter or to cation disorder in the albitic jadeite structure.

Discussion

Raman spectrum of jadeite: theory and reality. The Raman spectrum of jadeite is well-characterized from high-quality terrestrial specimens and density-functional theory calculations$^{19,20}$. It has strong characteristic A$_g$ symmetry peaks associated with the silicate chains at ~377, ~700, ~990, and ~1037 cm$^{-1}$ as well as bands associated with octahedral site vibrations at ~203, 328, and 528 cm$^{-1}$. The experimental Raman spectrum of tissintite is quite similar, though peak shifts relative to jadeite are large enough to observe a difference with strong peaks at ~377, 693, and 997 cm$^{-1}$, and with less intense peaks at ~203, 415, 523, and 573 cm$^{-1}$. The ~1037 cm$^{-1}$ peak is absent. These Raman features are consistent with both experimental and simulation results for a C2/c clinopyroxene (e.g., refs. 20–22), however, all the Raman bands of tissintite become progressively broader with increased substitution of (Si$^{4+}$+Mg$^{2+}$) for octahedral Al$^{3+}$ and increased cation and vacancy disorder on the M2 site$^{(Ma C. et al., 2015)}$. In supersilicic jadeites, such as albitic jadeite, the presence of octahedral Si is associated with the observation of new Raman peaks at ~340, 596, and 1110 cm$^{-1}$ (ref. 21).

We have computed theoretical Raman spectra for phases similar in structure with jadeite, but spanning a compositional range encompassing that observed in the three meteorites: ordered albitic jadeite (Na$_{0.75}$□$_{0.25}$)$^{M2}$(Al$_{0.75}$Si$_{0.25}$)$^{M1}$(Si$_2$)$^6$O$_{6}$, jadeite$^{50}$ albite$^{50}$ (Na$_{0.875}$□$_{0.125}$)$^{M2}$(Al$_{0.875}$Si$_{0.125}$)$^{M1}$(Si$_2$)$^6$O$_{6}$, jadeite, (Na)$_2$$^{M2}$(Al)$^{M1}$(Si$_2$)$^6$O$_{6}$, tissintite (Ca$_{0.75}$□$_{0.25}$)$^{M2}$(Al)$_{0.75}$$(Si_1.0)$$^{M1}$$^6$O$_{6}$, kushiroite (i.e., Ca-Tschermak pyroxene) (Ca)$^{M2}$(Al)$^{M1}$(Al$_{1.0}$Si$_{1.0})$$^{M1}$$^6$O$_{6}$, and silipyx (□)$^{M2}$(Si)$^{M1}$$^6$O$_{6}$ (Fig. 4B-G). The results of the jadeite simulation are in good agreement with the measured spectra for jadeite (RRUFF ID_R050220). Computed peaks are slightly shifted relative to measured frequencies, as is normally the case with density functional theory (DFT) calculations$^{22,23}$. The other spectra have similar patterns, typical of pyroxenes, and the differences among them can be quite subtle.

The computed spectrum of pure jadeite has a set of broad peaks at low frequencies, in three groups: 192, 195, and 210 cm$^{-1}$; 243, 273, 289, and 313 cm$^{-1}$; and the most intense peaks in this region at 353, 371, and 372 cm$^{-1}$, with a shoulder at 414 cm$^{-1}$. These peaks are dominated by various bending movements of tetrahedra acting as rigid bodies. M2 cations like Na and Ca participate in these low-frequency modes, but their presence is more significant in infrared-active modes in the same frequency range. In non-centro-symmetric structures, which develop due to ordering of vacancies, M2 cations contribute to the broadening of these sets of low-wavenumber peaks. The spectra of all the pyroxenes computed here share these three sets of peaks and their absolute and relative intensities are all similar. It is very unlikely that these peaks at below 450 cm$^{-1}$, whether computed or measured, can be used to reliably distinguish the cation proportions or to quantify the number of vacancies.

Next, there is then a broad weak peak due to the cation on the M2 site moving against the sublattice of oxygen atoms, centered at 502 cm$^{-1}$ in jadeite, at 591 cm$^{-1}$ in albitic jadeite, and at 648 cm$^{-1}$ in silipyx. This mode occurs in other high-pressure minerals with octahedral silica, like akimotoite (ref. 24) and post-perovskite, where it is the most intense mode (ref. 25). In pyroxenes, although the peak center is quite composition dependent, this mode is unlikely to be clearly observed experimentally due to its low intensity and diffuse peak shape.

The most intense Raman peak, typical of pyroxenes, is calculated at 572 cm$^{-1}$ in pure jadeite and at 638 cm$^{-1}$ in kushiroite. The addition of vacancies on the M2 site leads to a nearly linear upward shift in the frequency of this mode, to 701 cm$^{-1}$ in albitic
jadeite and 729 cm$^{-1}$ in silipyx. This mode is dominated by the breathing of the (M2)O$_6$ octahedra. At still higher frequencies one finds weak modes due to breathing of the SiO$_4$ tetrahedra; in some compositions these modes vanish almost completely (Fig. S3).

In summary, computed and observed Raman spectra show that the number of vacancies and the identity of the M1 cations influence peak positions and relative intensities. However, in practice, distinguishing between jadeite and related minerals using Raman spectra likely requires rather idealized observational conditions, with the laser focused onto a single crystal large enough to exceed the spot diameter (typically at least ~1-2 μm). In shocked meteorites, especially those shocked heavily enough to contain significant amounts of jadeite, the rapidly grown HP polymorphs typically occur mostly as tiny crystals or as aggregates of crystallites a few nm up to rarely more than ~2 μm in size. Furthermore, as a rule, HP phases often occur as intergrown composites of various polymorphs. As such, Raman spectra acquired from natural shock melt veins in meteorites are often of relatively poor quality, with broadened peaks, low intensities leading to missing peaks, and mixed-phase excitation. This means that the distinctive differences between true jadeite and defective pyroxenes like tissintite and albitic jadeite are challenging to observe. Indeed, there are a number of published cases where spectra with only a few (perhaps only one) of the main jadeite peaks and significant peak broadening are reported but have nevertheless been interpreted specifically as evidence of the presence of jadeite$^{26,27}$.

**Stability under electron beam irradiation.** As seen in previous studies, many minerals, especially those that are metastable at ambient conditions, quickly become amorphous when exposed to an electron beam at the current and voltage typically used in SEM and EPMA instruments (for example, bridgmanite; ref. 29). Stoichiometric jadeite is not beam-sensitive; it continues to yield high-quality EBSD patterns under extended irradiation. The two recently identified high-pressure vacancy-rich pyroxenes (i.e., tissintite and albitic jadeite) respond in different ways to the electron beam. Despite having about one-quarter of the M2 sites vacant, tissintite is not beam sensitive$^{14}$. On the other hand, albitic jadeite, which also has excess Si on the M1 site, is highly beam sensitive$^{4,5}$. If Si on the M1 site were the sole factor governing beam stability, all the albitic jadeite reported here would presumably show similar behavior, as all three cases have ≥0.10 Si on M1 site. However, the albitic jadeite in Ozerki is beam-stable; we collected quality EBSD patterns from this occurrence with no special beam exposure precautions (Figs. 6A, B). In contrast, the albitic jadeite in Chantonnay and Chug Chug 011 are both beam-sensitive; EBSD contrast disappeared within seconds of switching to a fixed beam spot (Figs. 6C, D). It seems, therefore, that the Ozerki occurrence of albitic jadeite is an exception to the Si-on-M1 rule for beam stability. We conclude that there must be additional factors governing behavior under the electron beam, but we do not yet have enough distinct cases to identify those additional factors. Possibilities include the effects of cation composition, site-ordering, or crystallite size.

**Solid-state transformation versus crystallization from a melt.** The formation of jadeite requires higher pressure than is found under static conditions anywhere in a chondritic parent body; the boundary of the reaction albite + SiO$_2$ rises from 1.65 GPa at 600 °C to 3.2 GPa at 1200 °C, where it meets the liquidus (ref. 29). Hence the occurrence of jadeite in a chondritic meteorite is considered *prima facie* evidence of an impact event strong enough to generate transient high pressure within the jadeite stability field. However, two categories of mechanisms have been proposed for the formation of jadeite: solid-state transformation and crystallization from a melt (ref. 30). Solid-state transformation is indicated when jadeite occurrences display one or more of the following textural features: 1) lamellar intergrowth with another phase such as lingunite, 2) coexistence with particle-like or stringer-like amorphous silica, 3) pseudomorphs after albite. We note that these textural categories may depend on the scale of observation; in the L6 ordinary chondrite Yamato 74445, for example, lamellar intergrowth of jadeite with lingunite is visible at 2,000× magnification and, in addition, inclusions of particle-like amorphous silica become apparent at 10,000× magnification$^{30}$. In contrast, jadeite growth from melt has been inferred from dendritic or spherulite-like textures. The dendritic development in Chelyabinsk includes needle-like and skeletal-rhombic crystals within feldspathic glass; the needles, in particular, are thought to have grown rapidly into feldspathic melt from nucleation sites on surfaces$^{31}$. The radial-concentric spherulite-like jadeite crystals in Novosibirsk, accompanied by chemical segregation of Na from K, are also inferred to indicate rapid crystallization from a melt$^{26,31}$. Furthermore, in both Chelyabinsk and Novosibirsk jadeite is found without any coexisting silica-rich phase (such as coesite, stishovite or glass), which is by itself a strong evidence against any solid-state transformation from feldspar. On the other hand, for albitic jadeite we cannot use the absence of a coexisting silica phase as evidence against solid-state transformation because at least some of the excess SiO$_2$ from the parent albite remains in the structure of this pyroxene.
In our observations of albitic jadeite, we find evidence for both solid-state transformation and crystallization from melt. In Ozerki, we have indications for both mechanisms within the same section: solid-state transformation is suggested by particle-like texture (Fig. 1A) and polycrystalline aggregates, whereas crystallization from melt is suggested by spherulite-like crystals in the interior of melt pools (Fig. 1B), with fine-grained dendritic crystallites at the rim of glassy pools (Figs. 1C, D, H), and skeletal hopper crystals (Fig. 1G). In Chug Chug 011, we see only dendritic texture, suggesting crystallization from a melt (Figs. 2A, B). In Chantonnay, the jadeite phase shows textures indicative of solid-state transformation: a lamellar texture at low magnification that resolves to small particles at higher magnification (Figs. 2C, D, 7).

**Fake news if not carefully characterized.** Before attempting to use the presence of a HP mineral to estimate P-T conditions in a shock-metamorphosed specimen, such a phase must be thoroughly characterized and its phase identification confirmed by the combination of one or more structure-sensitive analytical methods (such as Raman spectroscopy, EBSD, XRD, or TEM) and co-located compositional microanalysis (e.g., by EPMA). P-T inferences without such information could be in error either because the phase is misidentified or because its stability field depends on composition and may only have been experimentally calibrated for a particular end member.

We catalogued a total of 19 documented occurrences of jadeite and its relatives in ordinary chondrites (Table 1). These data include 13 type L6 meteorites, two type L5, and one each of types LL5, LL7, H5/6, and H6. Most of these reports combine Raman data with near-albite compositional analyses. Only in four meteorites (Château-Renard, NWA 8275, Villalbeto de la Peña, Yamato 791384; references in Table 1) has the mineral structure been verified by TEM. Most published reports do not provide documentation whether the pyroxene phase coexists with a silica phase. Only one study documents jadeite coexisting with coesite (NWA 8257; ref. 32). At least one published study has foregone compositional analysis and relied on Raman spectra alone. The danger of such an inference was already shown by the presence of omphacite in the L6 Château-Renard (ref. 33) with a Raman spectrum indistinguishable from jadeite. In such a case, an analysis showing near-albite composition can rule out omphacite, but the Château-Renard observation shows that Raman spectra may not uniquely distinguish jadeite from other sodic pyroxenes with stability fields different from those of pure jadeite.

The albitic jadeite in Ozerki and Chug Chug 011, both L6 chondrites, exhibits similar textures (mostly indicating crystallization from melt), yet Ozerki is classified as shock S4/5 (based on undulatory extinction, planar microstructures, and shocked melt veins) whereas Chug Chug 011 has been classified as S2. We dispute the accepted classification of shock stage Chug Chug 011, given our observations of mosaicism in olivine and various shock-related melt veins, including one containing albitic jadeite. The investigated thin section of Chug Chug 011 shows features more consistent with shock stage of at least S4. It remains to be determined, then, what shock pressure is implied by this mineral and its textural characteristics.

**P-T histories.** Shock-related melt veins (MVs) are generally thought to indicate local temperatures exceeding the liquidus of the matrix material, either under pressure or upon shock pressure release\(^{34}\). If the MV furthermore contains HP phases that grew from a melt, one may further infer that peak conditions were above the liquidus at HP and cooled enough for nucleation and growth of new phases to occur before pressure release. In such a case, constraints on the cooling time of a MV translate directly into a minimum time for the duration of the high-pressure pulse and hence the characteristics of the impact event. On the other hand, the interpretation of HP phases with textures indicative of solid-state transformation despite being found within melt veins is more ambiguous. In the Ozerki section, albitic jadeite with texture suggesting crystallization from the melt was found in the middle of MVs up to ~300 μm wide. The time for a 300 μm wide slab to cool from super-liquidus temperatures (~2000 °C) while surrounded by cool matrix (~100 °C) is estimated to be ~6.5 ms for using typical rock thermal diffusivity values\(^{35}\).

Concerning pressure, albitic jadeite is less dense than lingunite and is expected to form from an albite precursor at lower pressure. The absence of lingunite in the studied meteorite sections suggests maximum pressure below 21 GPa, the minimum pressure required for the formation of lingunite\(^{36}\). Other bounds on pressure are traditionally based on experimental data involving stable, stoichiometric pyroxenes coexisting with certain phase assemblages. For example, the experiments of Bobrov et al.\(^{37}\) show that a pyroxene of composition Jd\(_{70}\)Di\(_{30}\) begins to exsolve majorite garnet and stishovite above 13.5 GPa. Delay in nucleation of garnet and stishovite might in fact weaken the 13.5 GPa constraint\(^{38}\) but, more importantly, the static experiments do not address either the upper or lower pressure limits for the metastable formation of albitic jadeite. In the Chug Chug 011 section, we found an additional MV that shows a mixed xieite-chromite Raman spectrum at the rim of the vein and minor peaks of wadsleyite in the
center of the vein (Fig. 4C). Xieite implies $P$ conditions in the range 18-23 GPa at 1800-1950 °C (ref. 38) and wadsleyite suggests 13-22 GPa (depending on temperature and Fe content; ref. 33). There may have been a $P$ and $T$ gradient across this vein (Fig. 1B). The albitic jadeite itself, however, does not add any robust constraint to this pressure history at this time. Static equilibrium experiments at known $P$ and $T$ have so far not formed albitic jadeite as far as we are aware. As a general matter, lowering of the activity of jadeite component by the addition of albite and Ca-eskolaite components should expand the boundaries of the pyroxene liquidus field and allow growth over a larger pressure range than that for stoichiometric jadeite. To quantify this expansion and reveal the actual conditions for formation of albitic jadeite, however, it would be necessary to calibrate the shock pressures through a combination of dynamic experiments and observed coexistences with other pressure-sensitive indicator minerals and assemblages.

We report on three new occurrences of the recently described pyroxene species known as albitic jadeite (refs. 4,5) in the L6 chondrites Ozerki, Chug Chug 011, and Chantonnay. Each new occurrence was characterized by high-resolution SEM imaging, EDS and WDS compositional analysis, EBSD, and Raman spectroscopy. In addition, the Chantonnay material was further studied by FIB-TEM (Fig. 7). Like the omphacite in Château-Renard (ref. 33), all three new examples of albitic jadeite display Raman spectra that could be considered indicative of jadeite, yet each differs significantly from jadeite in their chemical composition. We note that most but not all albitic jadeite is beam-sensitive, and, unlike true jadeite, rapidly becomes amorphous during EBSD analysis and loses Na during TEM-EDS analysis. To substantiate the extent to which Raman spectra can differentiate among jadeite and related species, at least under ideal conditions, we used density functional theory and a model for thermal effects to compute the 300 K Raman spectra of the compositions jadeite (Jd), albite (Ab), Jd$_{50}$Ab$_{50}$, tissintite, and kushiroite, all in the C2/c clinopyroxene structure. While the computed spectra do show systematic differences, they are subtle enough to suggest that natural specimens can only be distinguished under ideal analytical conditions. Textural evidence shows that albitic jadeite may form both by crystallization from a melt and by subsolidus transformation; in the case of Ozerki, both mechanisms were seen within the same thin section. We caution that formation conditions for albitic jadeite and other clinopyroxenes that share Raman bands with jadeite have not been characterized experimentally or fully cross-calibrated with other mineralogical indicators of shock conditions. Hence, to avoid possible errors (probably overestimates) in minimum pressure constraints based on the presence of jadeite, it is necessary to characterize any meteoritic occurrence with sufficient compositional and structural information to confirm that it is true, near-stoichiometric jadeite and not one of the related but distinct pyroxenes.

**Methods**

**Sample characterization.** Ozerki fell on 21st June 2018 in Russia and the first samples were recovered only four days after the fall. It is an L6 chondrite, moderately to strongly shocked (S4/5), without signs of weathering (W0). Chug Chug 011 is a find, recovered in 2018 in Antofagasta, Chile; it is catalogued as L6, weakly shocked (S2; but see discussion in text), with minor signs of weathering (W1). Chantonnay is a historical fall (5th August 1812), classified as L6, moderately shocked (S4) and unweathered (W0)(refs. 16, 39). The largest known specimen is part of the Natural History Museum Vienna (NHMV, Austria) meteorite collection.

One polished section of Ozerki and of Chug Chug 011 were investigated. Two polished thin sections of Chantonnay (NHMV-N9836 and NHMV-M5621) from the NHMV were analyzed. As our focus was on shock effects, we mainly studied melt veins and melt pockets in a systematic search for shock-modified mineral grains and high-pressure polymorphs.

**Optical microscopy.** We used transmitted and reflected light microscopy to characterize color, texture, and likely mineralogy of phases large enough to be resolved optically.

**Scanning electron microscopy and electron backscatter diffraction.** We used two SEMs to characterize the texture and mineralogy of the meteorites. The Chantonnay sections were initially carbon-coated and investigated with a JEOL JSM-6610 LV instrument at the NHMV, equipped with a highly sensitive backscattered electron detector and an EDS. Analyses were conducted using a 15 kV accelerating voltage and a ~20 nA probe current, yielding analytical volumes with diameters less than 3 μm. Additional SEM analyses were performed at the California Institute of Technology (Caltech) GPS using a Zeiss 1550VP field-emission SEM equipped with an angle-sensitive backscattered electron detector, 80 mm² active area Oxford X-Max Si-drift-detector EDS, and an HKL EBSD system. SEM imaging and EDS analyses used a 15 kV accelerating potential and a 120 μm field aperture in high-current mode (~4 nA probe current), yielding imaging resolution better than 2 nm and an activation volume for EDS analysis ~1–2 μm$^3$ on
silicates. Single crystal EBSD analyses at sub-micrometer scale were performed at 20 kV and 6 nA in focused beam mode with a 70° tilted stage on uncoated specimens in "variable pressure" mode (25 Pa of N₂ gas in the chamber to reduce specimen charging). Imaging, mapping, semi-quantitative EDS analysis, and EBSD were conducted using the SmartSEM, AZtec, and Channel 5 software packages.

**Electron probe microanalyzer.** We used two JEOL JXA-8530F Field Emission Electron Microprobes, at the University of Muenster and at NHMV, for precise determination of the chemical composition of the studied minerals. Both instruments are equipped with five wavelength-dispersive spectrometers (WDS) and one EDS. Mineral analyses were performed with an accelerating voltage of 15 kV. For minerals, we used a 20 nA focused beam current, 20 s counting time on peak, and 10 s for each background position. For beam-sensitive minerals, we used a slightly defocused (5 μm diameter) beam, 5 nA probe current, and counting times of 10 s on-peak and 5 s on each background position. Natural mineral standards used were albite (Na, Si, Al), wollastonite (Ca), olivine (Mg), almandine (Fe), spessartine (Mn), orthoclase (K), rutile (Ti), chromite (Cr), and Ni-oxide (Ni) with ZAF matrix correction.

**Transmission electron microscopy.** We used a FEI Nova 600 Nanolab DualBeam focused ion beam (FIB) and scanning electron microscope (SEM) for the lift-out sample preparation. The sample thinning was finalized with an 8 kV 19 nA Ga-ion beam. The TEM analysis was performed on a FEI Tecnai TF20 with super-twin objective lens, operated at 200 kV. The EDS data were collected in TEM mode and scanning mode using a 70 μm C2 aperture and an EDAX SiLi detector. The data collection used 10 eV/channel and 51.2 μs process time, to achieve 50-500 cps signal and 20-50% deadtime. The FIB and TEM facilities are in the Kavli Nanoscience Institute at Caltech. The selected area electron diffraction (SEAD) patterns and EDS results were processed using Gatan DigitalMicrograph™ and FEI TEM Imaging & Analysis software.

**Raman Spectroscopy.** A dispersive confocal Renishaw inVia Reflex Raman microscope equipped with a 514 nm Ar-ion laser at the National Hellenic Research Foundation was used with a 100× objective lens. Spectra were collected in the Stokes region for Raman shifts from 200–1600 cm⁻¹. The laser beam was spread across ~1–2 μm spots at relatively low incident power (ca. 5 mW) to minimize sample damage. For each spot analysis we averaged spectra over 3 consecutive 60 sec accumulation times. Gaussian-Lorentzian peak fitting was used to remove background and estimate peak center frequencies. Collected spectra were compared with published data from the RRUFF database and the Handbook of Raman Spectra. The location of each Raman spot analysis was recorded so that co-located EPMA or SEM-EDS analytical points could be collected, allowing coupled structural and compositional characterization at common spots. Raman mapping was performed on a Renishaw InVia Confocal Raman microscope at the Mineral Spectroscopy Laboratory in Caltech. The 514 nm laser was set to <2 mW power to avoid laser damage. Each spectrum in the map was collected for 5 sec with 3000 line/mm diffraction gratings, corresponding to Raman shifts of 200-1100 cm⁻¹. Color maps were produced based on the integrated area of three diagnostic peaks over a linear baseline.

**Raman Computational Method.** We computed the theoretical Raman spectra of jadeite-related pyroxenes from the density-functional perturbation theory as implemented in the ABINIT package. The effect of the core electrons is taken into account using norm-conserving pseudopotentials. The electronic wavefunctions are described using planewaves, with kinetic energy cutoffs of 40 Ha (1 Ha = 27.2116 eV). The reciprocal space is sampled using a regular grid of k points. The computational methodology, including the pseudopotentials and the various parameters, was previously validated on various other minerals, and is similar to that used for the WURM project. Line broadening appropriate for thermal noise at 300 K was added to the line frequencies computed at 0 K.

**Declarations**

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Author contributions

I.B., with the help from P.D.A., J.H., C.M., L.F., designed this research. I.B., P.D.A., J.H., C.M., S.X., A.P., L.F., observed and analyzed the meteorite sections. R.C. performed and interpreted the first-principles calculations. All authors participated to writing and reviewing the manuscript.

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**Table**
Table 1 Summary of the occurrences of jadeite and its relatives in ordinary chondrites.

| Meteorite   | Type     | Applied Method | Chemistry                                      | Coexistence with SiO₂ | Likely formation mechanism |
|-------------|----------|----------------|-----------------------------------------------|------------------------|----------------------------|
| ALH81007318 | L6       | jd             | n.a.                                          | n.o.                   | n.p.                       |
| Chantonnay (this study) | L6       | jd             | ambiguous: jd or Ca-px | C2/c                  | SEM-EDS: Ca₀.0₃Na₀.₄₂Mg₀.₁₁Al₀.₉₃Si₂₀.₁₀O₆ | n.o.                     |
| Château- Renard33 | L6       | jd+Ca-px       | ambiguous: jd or omph                       | P2/n                  | EPMA: Ca₀.₀₇₋₀.₁₈Na₀.₂₅₋₀.₄₆Fe₀.₁₆₋₀.₁₉Mg₀.₃₅₋₁.₁₂Al₀.₂₉₋₀.₅₇Si₁₂₋₂₀.₄₉O₆ | n.o.                     |
| Chelyabinsk31 | LL5      | jd+ol          | n.a.                                          | n.a.                   | c.f.m.                     |
| Chug Chug 011 (this study) | L6       | jd             | n.a.                                          | n.a.                   | s.s.t.                     |
| GRV 05208226 | L6       | jd             | poor quality                                  | n.g.                   | n.p.                       |
| Novosibirsk26 | H5/6     | jd+opx+ol/jd+gl| n.a.                                          | n.a.                   | c.f.m.                     |
| NWA 827552  | LL7      | jd+coe/jd+amo  | Consistent with jd and coe                    |                        |                            |
| Ozerki (this study) | L6       | jd             | n.a.                                          | n.a.                   | c.f.m.                     |
| Peace River51  | L6       | +              | +                                             | +                      | gl                         |
| Pervomaisky45 | L6       | jd+maj-ppr+ol+Ca-px| n.a.                                          | n.a.                   | s.s.t.                     |
| Roosevelt Country 10652 | L6       | jd+lin+fsp     | n.a.                                          | n.g.                   | n.p.                       |
| Sahara 9822248 | L6       | jd             | n.a.                                          | n.a.                   | s.s.t.                     |
| Sixiangkou50  | L6       | jd+maj         | n.a.                                          | n.a.                   | n.p.                       |
| Taiban53      | L5       | jd+maj/jd+Ca-cpx| n.a.                                          | n.a.                   | c.f.m.                     |
| Villalbeito de la Peña46 | L6       | mixture of jd+lin| Consistent with lin                          |                        |                            |
| Y-7510047     | L6       | jd+mask        | n.a.                                          | n.a.                   | s.s.t.                     |
| Y-7444549     | L6       | lin+jd*        | n.a.                                          | n.a.                   |                            |
| Y-79138447    | L6       | jd             | jd (C2/c)**                                    | jd+amo                | EPMA: Ca₀.₀₉Na₀.₈₈K₀.₀₃Fe₀.₀₃Al₁₂₁₈Si₂₉₂₀O₈ | n.o.                     |
| Y-841055      | LL5      | +              | +                                             | +                      | n.p.                       |
| NWA 501156    | L6       | jd+maj         | n.a.                                          | n.a.                   |                            |

n.o.: not observed; n.a.: not applied; n.g.: not given; n.p.: not possible to infer because of absence of adequate images; s.s.t.: solid-state transformation; c.f.m.: crystallization from melt; RS: raman spectroscopy; EBSD: electron back-scatter diffraction; TEM: transmission electron microscopy.

Where more than one study reported jadeite and its relatives, we used the most recent one (citation number in bold). Abbreviations: jd: jadeite; omph: omphacite; maj-prp: majorite-pyroxene; lin: linungite; opx: orthopyroxene; Ca-px: high-Ca pyroxene; fsp: feldspar; coe: coesite; mask: maskelynite; gl: glass; amo: amorphous material.

* Ozawa et al. (Ozawa, et al., 2009) claim in the text (based on BSE images) the coexistence of linungite+jadeite, however, their Raman spectrum (their fig.7f) shows the characteristic strong peak for linungite only. **: applied method was synchrotron x-ray diffraction. + not sufficient data to
Figures

**Figure 1**

Back-scattered electron (BSE) images of occurrences of albitic jadeite in Ozerki. The images display textures indicative of both solid-state transformation (A) and crystallization from melt (B-F). A) Particle-like texture of amorphous material mixed with albitic jadeite. B) Spherulite-like crystals at the rim of a glassy melt pool. (C, D) Dendritic crystals showing a characteristic zonation inward from the melt-vein boundary. (E, F, G) High-resolution images of skeletal-hopper and (H) dendritic crystals inside a melt pocket.
Figure 2

BSE images showing textural features of albitic jadeite in Chug Chug 011 (A, B) and Chantonnay (C, D). (A) 12,000× view with locations of co-located Raman spots indicated. (B) 40,000× view of dendritic growth pattern. (C) BSE image of a melt vein in Chantonnay, with a melt pool at the center. (D) Enlargement of the melt pool within melt vein matrix. White circle denotes a Raman spot (spectrum A_HP03_bright area in Fig. 4D). The dashed box marks the location of the extracted FIB section (see Figure 7).
The silipyx \([\text{Spx}; (\square)M2(Si)M1(Si2)TO6]\) – jadeite \([\text{Jd}; (Na)M2(Al)M1(Si2)TO6]\) – kushiroite \([\text{Ku}; (Ca)M2(Al)M1(AlSi)TO6]\) diagram, showing notable compositional end-members albite (Ab), Ca-eskolaite (Ca-Esk), and anorthite (An), as well as jadeite, tissintite, and albitic jadeite analyses from the literature4,5,26,31-33,45-50 and from this work. Contours indicate atoms of Si per 6 O formula unit (long-dashed) and vacancies per formula unit (short-dashed). A) Published data. (B) This work. In (B), the open symbols correspond to Chantonnay analyses showing the effect of Na loss, which affects some beam-sensitive albitic jadeites.

**Figure 3**

**Figure 4**

Measured (A) and calculated (B-G) Raman spectra. All spectra are normalized to common maximum intensity and offset for clarity. A) Raman peaks at ~376, 698, 986, and ~1036 cm\(^{-1}\) are typical for jadeite (Jd; reference spectrum R050220 from RRUFF database shown). Tissintite (Tis), by contrast, has a single broad peak centered at ~1000 cm\(^{-1}\) (ref. 14). Spectra in Ozerki have all four main Raman peaks. In Chug Chug 011 (CC), numerous spectra display the peak at 698 cm\(^{-1}\) associated with jadeite, however the 376 cm\(^{-1}\) peak of jadeite is shifted to ~388 cm\(^{-1}\) and the two high-wavenumber peaks are not resolved. In one CC spectrum, the peak at 960 cm\(^{-1}\) indicates an apatite inclusion. In Chantonnay (Ch), the peaks in at ~204, 222, 254, 377, 699, 987, and 1038 cm\(^{-1}\) are all near-ideal matches for jadeite. In calculated Raman spectra for (B) kushiroite \([\text{(Ca)}M1(Al)M2(Al, Si)TO6]\), (C) tissintite \([\text{[(Ca0.75□0.25)M1(Al)M2(Al, Si)TO6]}]\), (D) jadeite \([\text{[(Na)M1(Al)M2(Si2)TO6]}]\), (E) intermediate \([\text{[(Na0.875□0.125)M1(Al0.875Si0.125)M2(Si2)TO6]}]\), (F) albitic jadeite \([\text{[(Na0.75□0.25)M1(Al0.75Si0.25)M2(Si2)TO6]}]\), and (G)
Raman mapping of feldspathic pool in Ozerki (A–C) and Chug Chug 011 (D–E). A) BSE image of the feldspathic pool, showing elongated and equant crystals at the edge and interior of the pool, respectively. B) Raman map (of the boxed area in the SEM image) superposed on reflected light image in greyscale. Red and blue color represent the intensity of diagnostic peaks of 'jadeite' and diopside, respectively. Purple color at the edge of the pool indicates coexistence of the two peaks. C) Representative Raman spectrum from the purple area. No Raman signature of polymorphs of silica is observed in this area. D) BSE image of Chug Chug 011 with mapped area indicated by white square. E) Raman maps of intensity of diopside peak in blue and 'jadeite' peak in red. Grey background indicates no strong fluorescence with no diagnostic peaks, corresponding to feldspathic glass.
Figure 6

Electron backscatter diffraction (EBSD) patterns. Albitic jadeite from (A, B) Ozerki and (C, D) Chug Chug 011, both indexed with a C2/c clinopyroxene structure. These materials were sufficiently beam-stable to collect a pattern.
Figure 7

Transmission electron microscopy data. (A) A focused ion beam (FIB) section extracted from the boundary of the Chantonnay melt pool at the location indicated in figure 2D. (B) Bright field image of the Si-pool end of the FIB section. The homogeneous matrix is Si-rich glass. The grains with contrast to the matrix are fine clinopyroxene (cpx) crystals. The different apparent brightness of the grains results from different crystallographic orientations; the darkest grains are viewed nearly along a major zone axis. (C) Selected area electron diffraction (SAED) patterns for three of the dark crystals seen in (B), rotated to low-index zone axes. All the patterns indicate a C2/c clinopyroxene structure.

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- Supplementarydata.xlsx
- Table1.docx
- SupplementaryInformation.docx
- SupplementaryInformation.docx