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Electrical discharge triggers quasicrystal formation in an eolian dune

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We report the discovery of a dodecagonal quasicrystal $Mn_{72.3}Si_{15.6}Cr_{9.7}Al_{1.8}Ni_{0.6}$—composed of a periodic stacking of atomic planes with quasiperiodic translational order and 12-fold symmetry along the two directions perpendicular to the planes—accidentally formed by an electrical discharge event in an eolian dune in the Sand Hills near Hyannis, Nebraska, United States. The quasicrystal, coexisting with a cubic crystalline phase with composition $Mn_{68.9}Si_{19.9}Ni_{7.6}Cr_{2.2}Al_{1.4}$, was found in a fulgurite consisting predominantly of fused and melted sand along with traces of melted conductor metal from a nearby downed power line. The fulgurite may have been created by a lightning strike that combined sand with material from downed power line or from electrical discharges from the downed power line alone. Extreme temperatures of at least 1,710 °C were reached, as indicated by the presence of SiO_2 glass in the sample. The dodecagonal quasicrystal is an example of a quasicrystal of any kind formed by electrical discharge, suggesting other places to search for quasicrystals on Earth or in space and for synthesizing them in the laboratory.

fulgurite | electrical discharge | quasicrystal | shock | solar system

Significance

The article presents the quasicrystal created by an electrical discharge, in this case accidentally created by a lightning strike or a downed power line in a wind-created dune in the Sand Hills of north central Nebraska. The discharge produced extreme temperatures (>1,710 °C) that led to the formation of a fulgurite, a tube of fused and melted sand along with traces of melted conductor metal from the power line. Within the fulgurite was found a "dodecagonal quasicrystal" composed of equally spaced atomic layers, each with 12-fold symmetry and quasicrystalline order that is impossible for ordinary crystals. The discovery suggests mechanisms for forming quasicrystals in nature (on Earth and in space) and in the laboratory.

An electrical discharge event in 2008 in the eolian dunes of the Sand Hills in the High Plains of north central Nebraska led to the accidental creation of a dodecagonal quasicrystal with a heretofore unreported composition $Mn_{72.3}Si_{15.6}Cr_{9.7}Al_{1.8}Ni_{0.6}$. The discharge, which may have been due to a lightning strike or to a downed power line, produced extreme temperatures (>1,710 °C) that led to the formation of a fulgurite composed of fused and melted sand together with traces of melted conductor metal from the power line (1). Within the fulgurite, the quasicrystal was found associated with a cubic crystalline phase with composition $Mn_{68.9}Si_{19.9}Ni_{7.6}Cr_{2.2}Al_{1.4}$, where the subscripts represent atomic percent.

Quasicrystals are solids with quasiperiodic translational order and rotational symmetry that are impossible for periodic crystals (2). In the case of a dodecagonal quasicrystal, the atomic structure consists of equally spaced atomic layers, each of which has crystallographically forbidden 12-fold symmetry. Due to their unconventional symmetries, quasicrystals have physical properties (elastic, electronic, photonic, phononic, friction, etc.) that are distinct from those of crystals and amorphous solids (3, 4), which makes them of interest for applications. The first reported example of a quasicrystal of any type is the spin-quenched icosahedral Al_6Mn alloy described by Shechtman et al., in 1984 (5). Until 2009, the only known quasicrystals were all synthesized in the laboratory using a variety of methods, in addition to melt-spinning, including slow quench from a liquid state followed by annealing, mechanical alloying, sputtering, vapor deposition, and thermal spraying (3, 6).

The search for quasicrystals in nature (7, 8) was motivated in part by the notion that they could point to compositions and formation methods that have not been considered previously. Indeed, the discovery of icosahedral $Al_{63}Cu_{24}Fe_{13}$ (8) and two other quasicrystal phases (9, 10) in the Khatyrka CV3 carbonaceous chondritic meteorite (11, 12) and the evidence that they formed in a high-velocity impact shock reaching temperatures of about 1,200 °C and pressures greater than 5 GPa (13, 14) inspired a series of laboratory impact shock experiments (15–18) that recreated the discovered natural quasicrystals as well heretofore unknown quasicrystalline phases.

The success of these experiments suggested that shock synthesis may be advantageous in producing quasicrystals with a multiplicity of elements (16), an idea which is currently being explored. It also inspired a search of red trinitite remnants produced by the fusion of arkosic sand and copper cables resulting from the high-pressure shocks and elevated temperatures generated by the Trinity nuclear test in Alamogordo, New Mexico, in 1945. That study led to the discovery of a previously unknown composition of icosahedral quasicrystal, $Si_{61}Cu_{30}Ca_7Fe_2$ (19).

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The current investigation was designed to explore a different possible nature-inspired mechanism for generating quasicrystals: electrical discharge. The discovery of a quasicrystal in a fulgurite with rarely observed 12-fold symmetry and a not been reported previously composition indicates that this approach may also be promising in the laboratory.

Results

Description of the Sample. The studied fulgurite sample is white in incident light with a metallic fragment visible at the end of the tubular sample (Fig. 1 *A* and *B*). The portion of the sample enlarged in Fig. 1*B* has been embedded and polished (Fig. 1*C*) and studied by scanning electron microscopy (SEM). The metallic fragment is mainly a melted Al alloy (originally a piece of Al-1350 alloy) with some regions more enriched in silicon and others more enriched in iron (*SI Appendix*, Fig. S1), but the mean composition can be written as $Al_{70}Fe_{16}Si_{10}Cu_2Cr_1Mn_1$ (Table 1).

The Al alloy is embedded in a silicate glass, mostly formed from melted quartz, which sets a temperature of >1,710 °C for its surroundings. There are also Ca, Na, and K components (Table 1). Electron backscatter diffraction (EBSD) data reveal that the Al alloy crystallizes in the *Pm*-3 $Al_{57}Mn_{12}$-type structure similar to that described (20) from the Variscan metagranitoids in Upper Carinthia, Austria (20).

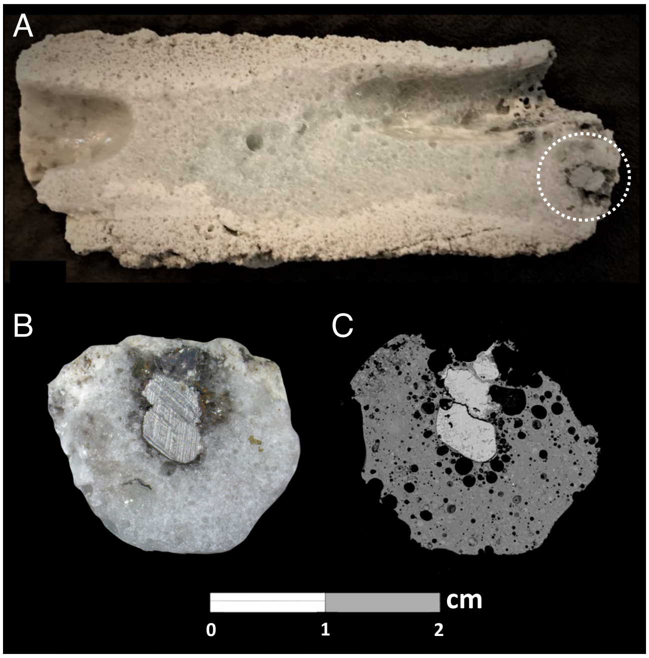

Fig. 1. (*A*) The Sand Hills fulgurite. The white dashed circle indicates the region enlarged in (*B*). (*C*) Panoramic SEM-BSE image of the enlarged region in (*B*).

At the contact between the Al metal alloy and the fused silicate glass there are transition regions (*SI Appendix*, Figs. S2 and S3) showing redox reactions between the two components. In one of the vesicles of the Al metal alloy (Fig. 2 *A* and *B*), there is a small brighter metallic portion, which is composed of two different phases (Fig. 2 *C* and *D*). The acicular grains have a mean composition of $Mn_{72.3}Si_{15.6}Cr_{9.7}Al_{1.8}Ni_{0.6}$, while the coexisting phase is $Mn_{68.9}Si_{19.9}Ni_{7.6}Cr_{2.2}Al_{1.4}$. There is some evidence of preferred orientations of the acicular grains, but the alignment is not precise enough to demonstrate a topotactic relationship with the host phase. EBSD data on the two phases showed that the first exhibits a crystallographically forbidden 12-fold symmetry (*SI Appendix*, Fig. S4), while the second is the cubic space group $P2_13$, with the Au_4Al-type structure (*SI Appendix*, Fig. S5).

To further characterize these materials, a thin portion of the Mn-Si-Cr-Al-Ni fragment was extracted by means of the focused ion beam (FIB) technique and studied with a transmission electron microscope (TEM). The TEM study revealed that, at the submicron length scale, the aggregate indeed consists of the two phases found with the SEM (*SI Appendix*, Fig. S6), where each individual grain is chemically homogeneous. The cubic $P2_13$ phase was confirmed by electron diffraction (*SI Appendix*, Fig. S7). A selected area electron diffraction (SAED) pattern along the 12-fold axis is shown in Fig. 3*A*. The pattern, consisting of sharp peaks arranged in an incommensurate lattice with 12-fold symmetry, is the characteristic signature of a dodecagonal quasicrystal. The high-angle annular dark-field (HAADF) scanning transmission electron microscopy (STEM) image in Fig. 3*B* shows that the real space structure consists of a homogeneous, quasiperiodic, and 12-fold symmetric pattern composed of dodecagonal clusters (*SI Appendix*, Fig. S8). Together, these

Table 1. Chemical composition (TEM-EDS and SEM-EDS data in atomic %) of the dodecagonal quasicrystalline phase, the normal phase coexisting with the quasicrystal, the Al-rich metal embedding the quasicrystal, and the silicate glass

	Quasicrystal					Cubic Phase			
	TEM-EDS	TEM-EDS	TEM-EDS	SEM-EDS	SEM-EDS	TEM-EDS	TEM-EDS	SEM-EDS	SEM-EDS
Al	1.7	2.0	2.0	1.4	1.7	1.7	1.5	1.3	1.2
Si	15.5	15.8	15.3	15.6	15.8	17.3	21.0	20.4	20.7
Cr	9.7	9.8	9.7	9.5	9.8	2.4	2.3	2.0	2.2
Mn	72.4	71.8	72.5	72.6	72.0	71.0	68.0	67.9	68.8
Ni	0.7	0.6	0.5	0.9	0.7	7.6	7.2	8.4	7.1

	Al-rich phase					Glass	
	SEM-EDS	SEM-EDS	SEM-EDS	SEM-EDS		SEM-EDS	SEM-EDS
Al	69.6	70.5	69.4	68.2	Na_2O	-	2.1
Si	10.2	10.2	10.4	11.8	K_2O	-	2.5
Cr	0.3	1.8	1.9	1.5	CaO	-	3.5
Mn	0.6	0.5	0.5	0.5	Al_2O_3	-	15.1
Fe	17.0	15.2	16.1	15.3	SiO_2	100.0	76.8
Cu	2.3	1.9	1.9	2.7			

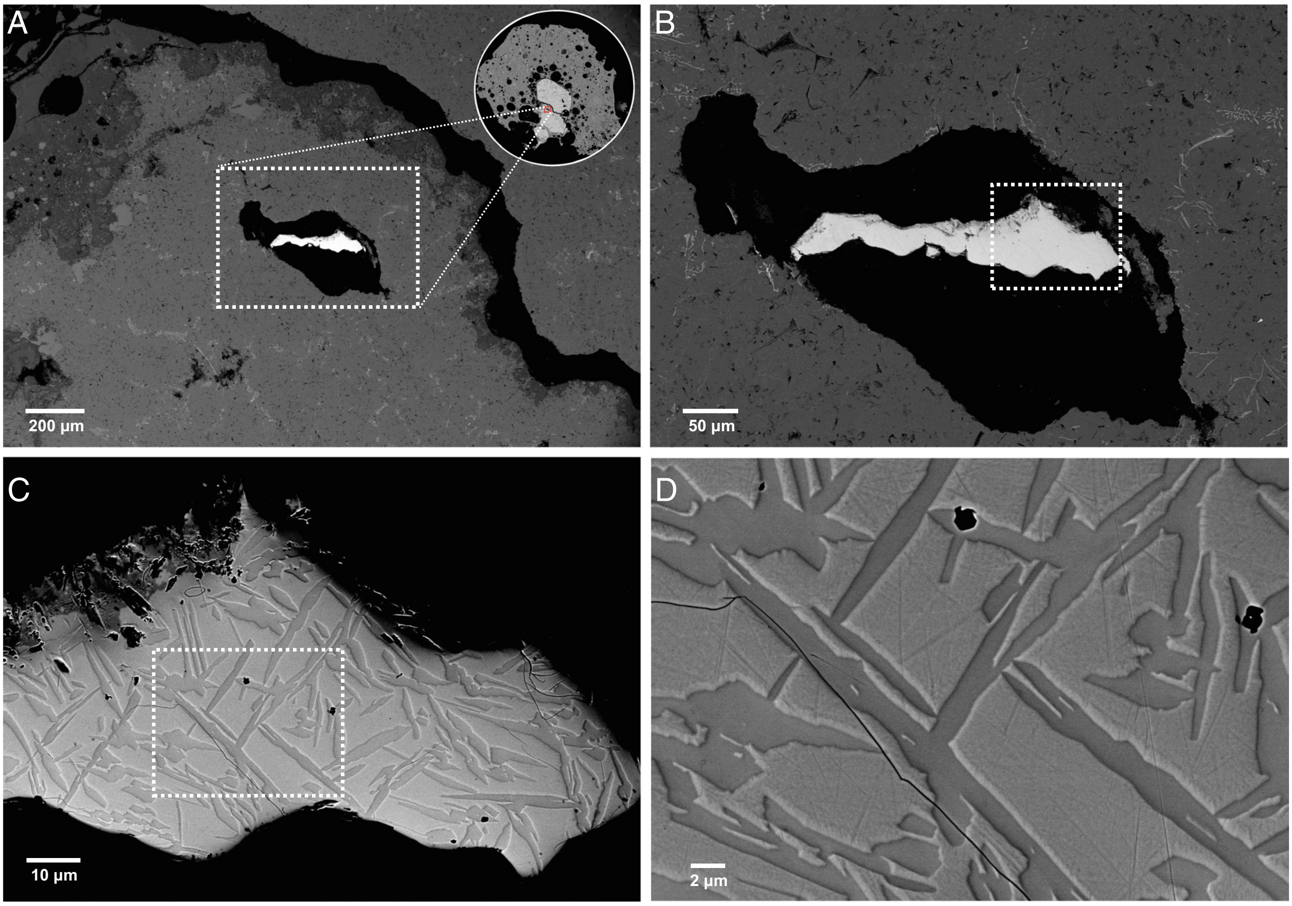

Fig. 2. (*A*) SEM-BSE image of a part of the metallic portion of the Sand Hills fulgurite given in the inset. The dashed white rectangle indicates the region shown in panel (*B*). (*C*) Grain containing quasicrystalline portions (acicular darker fragments) and crystalline portions. The two portions are highlighted in (*D*).

TEM/STEM results provide conclusive evidence of quasiperiodic translational order and crystallographically forbidden dodecagonal symmetry in a material formed by electrical discharge.

The dodecagonal quasicrystal also coexists with small portions of the hexagonal *P*6/*mmm* approximant observed in Mn-(V,Cr)-Si systems (21) similar to other dodecagonal quasicrystals that have been found to coexist with approximant crystals composed of the same local structural units as in the quasicrystal (22). As shown in *SI Appendix*, Fig. S9, there are portions that can be described as periodic and exhibit pseudo–12-fold symmetry (see *Inset* in *SI Appendix*, Fig. S9). According to Iga et al. (21), the structure of the hexagonal approximant can be described as the tiling of two types of local structural units, namely an equilateral triangle and a square with the same edge length of 4.57 Å, which correspond to the A15-type and Zr_4Al_3-type structures in three dimensions, respectively.

Origin of the Sample. The Sand Hills fulgurite studied here (1) was found following a storm at the site of a downed power line. Because the event was not definitively associated with an observed lightning strike, the roles of lightning and the power line in forming the fulgurite are unclear. The fulgurite was about 2 m long along its main channel and had a maximum diameter of 8 cm. Several smaller channels (2 cm or smaller in diameter) branched off this main channel. The fulgurite was mostly oriented horizontally with respect to the ground, but it did penetrate to ~10 cm depth at a maximum.

The fulgurite consisted primarily of fused and melted sand. The sand making up the dunes in the Sand Hills is composed primarily of quartz and feldspar, with modal mineralogy estimates showing 50 to 80% quartz (23–24). K-feldspar is present in about 5 to 15% (25), and plagioclase makes up the remainder. This mineralogy is reflected in the normative composition of the silicate fulgurite glass, which includes about 85% quartz, 5% K-feldspar, and 10% plagioclase (Table 1). Accessory minerals (<2%) in the source sand include mica, amphiboles, pyroxenes, epidote, zircon, and tourmaline (26). Grain sizes are typically ~200 μm (23). The source of the Sand Hills eolian deposits may be the Ogallala Group [Miocene to Pliocene (27)] or other Pleistocene alluviums (28). The dunes in the Sand Hills formed in the Holocene [5,000 to 8,000 y before present (29)].

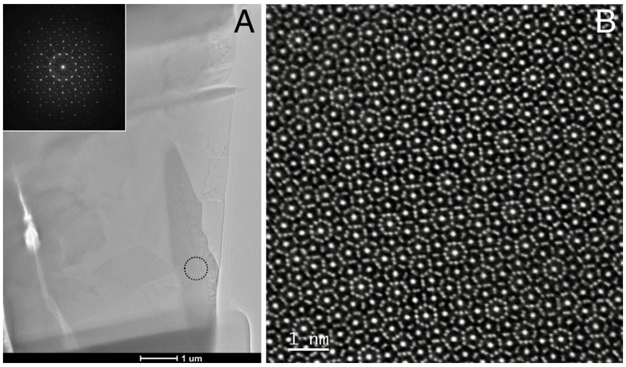

Fig. 3. TEM/STEM data obtained on the FIB lamella. (*A*) The black circle in the acicular, quasicrystalline grain indicates the region where the electron diffraction pattern (*Inset*) has been collected. (*B*) A HAADF-TEM image of a portion of the quasicrystalline grain.

The fulgurite also contained bits of anthropogenic metal conductor from the power line. Power line conductors in the United States are composed of either copper or aluminum, with aluminum being the material of choice for most modern conductor lines due to the significantly lower cost of Al vs. Cu. Aluminum power lines are composed of Al-1350, an alloy consisting of 99.5% Al, 0.4% Fe, and Si, Cr, Mn, Cu, and Zn (together ~0.1%, see ref. 30). Aluminum power lines are also often reinforced with a galvanized steel core (31), especially over rural areas (such as Hyannis, Nebraska), where the steel usually makes up about a third of the mass of a power line (32).

The Mn for the new quasicrystal may have originated from the power line wire (most probably from the steel core of the wire, which is required by established standards to have >0.5 wt.% Mn) followed by dissolution of iron into the aluminum metal, leaving the Mn-rich phase behind. Elements such as Mn, Cr, Ni, Fe, and Al have very different redox potentials and can be strongly fractionated into different phases during transient melting events (15). Alternatively, the Mn may have been present as part of the heavy mineral inventory of the sand (for instance, as hausmannite, Mn_3O_4), which would have formed metal as the oxides were reduced (e.g., $Mn_3O_4 + 8/3\ Al = 3\ Mn + 4/3\ Al_2O_3$, $\log K = 10^{30}$ at 1,000 °C). Winspear and Pye (33) reported MnO at ~0.03 wt.% of the Sand Hills sediment. The acicular Mn-rich grains are volumetrically a small fraction of the fulgurite ($<10^{-4}$), and the supply of Mn would be sufficient with either the conductor metal or the sand as sources.

Pasek and Pasek (1) argued that the fulgurite was formed solely by the downed power line based on the presence of melted conductor wire and cristobalite within the fulgurite. Cristobalite was specifically used as an indicator of an artificial discharge source as its formation from pure SiO_2 requires elevated temperatures (~1,500 °C) persisting for >600 s based on the calculations by Breneman and Halloran (34). Lightning-induced heating to these temperatures is expected to persist for much shorter timescale due to heat dissipation by diffusion [~0.3 s (35)], whereas longer-heating timescales are plausible for artificial sources.

However, lightning remains a possible cause because, as emphasized by Elmi et al. (36, 37), cristobalite formation can be greatly accelerated by the presence of K-feldspar and plagioclase, which were observed in the Sand Hills fulgurite, allowing crystallization from a dry eutectic melt rather than the subsolidus SiO_2 phase transformation considered in the analysis of Breneman and Halloran (34). The additional components act as a flux to lower the melting temperature of pure silica such that cristobalite may precipitate over a range of temperatures as the melt cools. Furthermore, cristobalite can remain as a metastable phase at temperatures 300 to 400 °C lower than its equilibrium formation temperature (38). The situation differs from natural granitoid minimum melting, where the presence of water lowers the eutectic temperature into the stability field of quartz.

In addition to the mineralogy of the Sand Hills fulgurite being consistent with formation by lightning, the Sand Hills fulgurite also extended well beyond the contact point with the aluminum wire and had a hardened tube-like morphology similar to conventional lightning-created fulgurites. In contrast, fulgurites formed by downed power lines typically form radially outward from the contact point of the power line with the ground and are friable (1). However, if the Sand Hills fulgurite formed by lightning, the lightning strike would have to have been near to where the downed cables touched the ground in order to incorporate and fully enclose the aluminum conductor in the glassy fulgurite.

Another consideration is pressure. The fulgurite may have experienced high-pressure conditions that coincided with the formation of the quasicrystalline material as lightning strikes are accompanied by pronounced pressure increases (15, 39–40). It is known that events involving transient high-pressure pulses can lead to the synthesis of quasicrystals (13, 15–19). However, all such transient high-pressure events release to longer-lived, low-pressure, high-temperature conditions. The presence of cristobalite clearly demonstrates lower-pressure conditions during the formation of this fulgurite. It remains unclear whether the quasicrystals observed in materials subjected to such pressure–temperature–time histories nucleated and grew exclusively during the high-pressure pulses and, indeed, whether the high-pressure pulse is necessary for their synthesis.

Discussion

To date, dodecagonal quasicrystals are rare compared with quasicrystals with decagonal symmetry (periodic atomic layers with quasiperiodic translational order and 10-fold symmetry) or three-dimensional icosahedral symmetry (3), although examples have been known since the Ni–Cr particles with 12-fold symmetry reported by Ishimasa et al. (41). Later, Iwami et al. (22, 42) synthesized grains of a Mn-rich dodecagonal quasicrystal with composition $Mn_{74}Si_{15}Cr_{10}Ni_1$ in a matrix of β-Mn-type $Mn_{73}Si_{17}Ni_7Cr_3$ cubic crystals by an elaborate and a highly controlled procedure that included alloying high-purity samples of the four elements, sealing samples of the ingot in silica, annealing at 700 °C for at least 130 h, and then water cooling.

By comparison, the dodecagonal quasicrystal described in this paper was formed by a completely uncontrolled process, lasting perhaps a few minutes, from a heterogeneous mix of natural and anthropogenic components and yet resulting in a similar intergrown morphology of crystal and quasicrystal and in quasicrystal composition close to the laboratory one in that it includes a fifth component, Al, that was not included in the laboratory synthesis. Just as the discovery of natural quasicrystals in the Khatyrka meteorite (8) pointed to the idea that shock synthesis may be an effective mean of searching for new elemental compositions that form multicomponent quasicrystals (16), the discovery of a dodecagonal quasicrystal formed by a lightning strike or downed power line suggests that electric discharge experiments may be another approach to be added to our arsenal of synthesis methods.

The results may also provide a clue to a remaining mystery regarding the exotic metallic alloys in the Khatyrka meteorite, namely exactly how and when they formed. There has already been reported ample evidence that high-velocity impact shocks played a critical role, including the observations of melting textures that show the nucleation and growth of quasicrystals (14) and other indicators that Khatyrka underwent a series of collisions (11, 43) over a period of hundreds of millions or perhaps billions of years. However, the impacts alone do not explain the highly reducing conditions needed to explain the presence of metallic Al in the quasicrystals and several of the crystalline phases found in Khatyrka grains. In their pioneering study of a glassy fulgurite, Essene and Fisher (44) noted that extreme reduction conditions and high temperatures could occur as a result of electrical discharges (i.e., lightning strikes) in the solar nebula from dust or impacts. The results presented here, together with the trace element abundances measured in natural quasicrystals (45), open the possibility that electric discharge in the early solar nebula may have played a key role that not only accounts for the requisite reducing conditions but also promotes quasicrystal formation.

Materials and Methods

The Sand Hills fulgurite described in this paper and shown in Figs. 1 and 2 was provided by one of the authors (M.A.P.). A portion of the sample (white dashed circle in Fig. 1*A*) was embedded in epoxy resin and prepared as a polished thick section (Fig. 1 *B* and *C*). The results presented here are from SEM and TEM techniques.

SEM. The instruments used were a Zeiss EVO MA15 Scanning Electron Microscope coupled with an Oxford INCA250 energy-dispersive X-ray spectrometer (EDS), operating at 25 kV accelerating potential, 500 pA probe current, 2,500 cps as average count rate on the whole spectrum, and a counting time of 500 s, and a ZEISS 1550VP field emission SEM with an HKL EBSD system, operated at 20 kV and 6 nA in focused beam mode with a 70° tilted stage and in a variable pressure mode (25 Pa) for EBSD analysis. Samples were sputter-coated with a 20-nm-thick carbon film for BSE imaging and EDS analysis.

TEM. In preparation for the study with TEM, thin lamellae from the fulgurite sample (Fig. 2) were prepared by FIB cutting using a Helios NanoLab G3 UC dual-beam FIB/SEM system. Sample thinning was accomplished by gently polishing the sample using a 2-kV gallium ion beam in order to minimize surface damage caused by the high-energy FIB.

Conventional TEM imaging, SAED, atomic-resolution HAADF-STEM imaging, and energy-dispersive X-ray spectroscopy (EDS) mapping were performed on a Titan Cubed Themis 300 double Cs-corrected STEM equipped with an extreme field emission gun source operated at 300 kV with a super-X EDS system. The system was operated at 300 kV.

Data, Materials, and Software Availability. All study data are included in the article and/or *SI Appendix*.

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