



Article

Constraints on iron silicide formation from a fulgurite from Southwick, Massachusetts, USA

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Abstract

Fulgurites are natural glasses that form when lightning strikes sand, soil, or rock and fuses the individual grains together to generate what is usually a tubular structure that follows the path of the strike. During this process, localised reducing conditions are conducive to forming rare minerals including iron silicides. This paper examines a fulgurite formed in Southwick, Massachusetts, USA, which displays an iron silicide that has a clearly defined reaction rim. The reaction rim demonstrates the production of a more silicon-rich rind consisting of Fe_5Si_3 on a core of Fe_2Si , and the most likely route to forming this material is by reaction of silicon gas with Fe_2Si at high temperature ($>1000^\circ\text{C}$), with a reaction timescale of about one second. This reaction suggests the high temperature, reducing conditions of a lightning strike favour reactions of condensed matter (e.g. liquid or solid iron minerals) with gas that occurs rapidly during the lightning strike. The conditions necessary to form these minerals suggest that the fulgurite became more reducing over time, as more Si entered the solid phase, perhaps as oxygen left the system, either as CO_2 or from the breakdown of SiO_2 gas.

Keywords: fulgurites; silicides; reduced minerals; reduction

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Introduction

Lightning is a ubiquitous natural process, and an individual strike can produce a peak power of 10^{13} W and a peak temperature of >30000 K all within about a millisecond (Kridner *et al.*, 1968). When lightning strikes soil, sand, or rock, the resulting rapid change in temperature can lead to the formation of a natural glass known as a fulgurite. This temperature change consists of an extremely rapid temperature increase due to the formation of the plasma channel (<1 second), followed by a slower, but still relatively rapid decrease as the heat dissipates conductively ($\sim$ minutes). A fulgurite is a glassy and usually tubular structure that follows the path of the strike. Fulgurites can be natural or artificial in origin. Artificial fulgurites are formed when a downed electric power line discharges into soil, typically over periods of minutes to hours. In contrast natural fulgurites form under much shorter timescales (<1 second) with much higher power (megawatts to gigawatts) (Pasek and Pasek, 2018).

Lightning strikes produce extremely high temperatures and through this rapid deposition of energy are capable of generating a highly reducing local environment (Sheffer, 2007). Due to the reductive environment formed when lightning strikes a material, Hess *et al.* (2021) argued that fulgurites could have played a role in supplying a source of reactive reduced phosphorus on

the early Earth surface. Fulgurites were one of the first modern rock types in which reduced phosphorus was discovered (Pasek and Block, 2009), probably present as both phosphides and phosphites, including solid CaHPO_3 (Bindi *et al.*, 2023a). The process of fulgurite formation (high temperature, high energy and highly reducing) leads to the formation of rare materials such as iron silicides, phosphides, phosphites (Pasek *et al.*, 2012), and even more complex alloys (Bindi *et al.*, 2023b).

Iron silicides are a group of rare minerals that form under reducing conditions. Silicides have been invoked as a possible solution to the presence of a light element in the Earth's core (Lin *et al.*, 2002; Kuwayama *et al.*, 2009; Hsieh *et al.*, 2020), and in general are rare in surficial rocks. Essene and Fisher (1986) were the first to investigate iron silicides in fulgurites, specifically the Winans Lake fulgurite, Michigan, USA. The Winans Lake fulgurite contains metallic spherules inside a glassy matrix, consisting of intergrowths of phosphides, metals and silicides. The silicides included compositions such as Fe_3Si_7 , FeSi and FeTiSi_2 . The Winans Lake fulgurite displays no signs of the spherules mixing with the matrix leading to the conclusion that the spherules were contemporaneous with the formation of the fulgurite. The cause of silicate reduction within the fulgurite was attributed to concurrent carbon oxidation, probably produced by combustion of a nearby tree root, as evidenced by the presence of graphite within the fulgurite.

Since Essene and Fisher (1986), there have been several more reports of iron silicides in fulgurites (see review by Feng *et al.*, 2021). Iron silicide minerals include fersilicite/naquite (FeSi), ferdasilicite/linzhiite (FeSi_2), gupeite (Fe_3Si), hapkeite (Fe_2Si),

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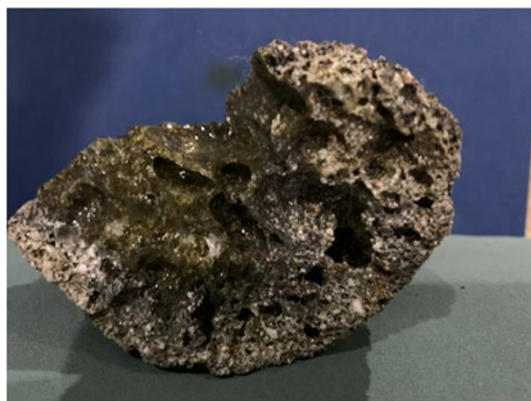
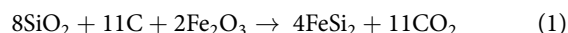


Figure 1. Image of the fulgurite from Southwick, Massachusetts, USA in cross section (specimen in private collection). Width is 7 cm.

luobusaite ($\text{Fe}_{0.84}\text{Si}_2$), xifengite (Fe_5Si_3) and suessite [$(\text{Fe}, \text{Ni})_3\text{Si}$]. Several of these have been found in fulgurites (Essene and Fisher, 1986; Sheffer and Dyar, 2004; Ramírez-Cardona *et al.*, 2006; Block, 2011; Pasek *et al.*, 2012; Roberts *et al.*, 2019; Bindi *et al.*, 2023a). The formation of iron silicides within fulgurites probably depends on the presence of reductants such as organic carbon to drive the removal of O from SiO_2 , though Sheffer (Sheffer, 2007) argued that carbon oxidation alone could not provide enough of a reducing environment to form iron silicides in all observed instances.

Iron silicides have been produced industrially for over 125 years to remove oxides from melted steel during casting processes (Brack *et al.*, 2015; Chepkasov *et al.*, 2020; Rappenglück, 2022). Industrially, the production of FeSi_2 follows the reaction:



An alternative route to production of iron silicides utilises a process called electrodeposition. (Hovestad *et al.*, 1997; Popovici *et al.*, 2005; Liang *et al.*, 2006; Saidin *et al.*, 2011; Delekar, 2022).

Building on the generation of silicides from industry, and noting the formation of silicides within some rocks, here we present data on an iron silicide within a Type II (Pasek *et al.*, 2012) fulgurite from Southwick, Massachusetts, USA. We investigate a particular occurrence of iron silicides in this fulgurite to better constrain how

these materials form under geologic settings, and especially in a lightning strike.

Methods and materials

The Southwick Massachusetts fulgurite (Fig. 1) was obtained from the finders who sold the sample on eBay to one of the authors (M.A.P.) in 2015. The finders described the location only by the nearest town where it was found (See Fig. 2), hence we will discuss the general geology of the Southwick area of Massachusetts (Schnabel, 1973), and contrast this sample with another nearby fulgurite to confirm its origin site. According to the Southwick geological quadrangle, the surficial area of Southwick is predominantly composed of reddish brown or light olive grey till forming on sediments dated to the Triassic (252–201 million years ago). The fulgurite was said to have formed in 2010 after a thunderstorm.

The proximity of the Southwick fulgurite to another fulgurite discussed by Sheffer *et al.* (2003) in Farmington, Connecticut (30 km) is noted. The target material of the Farmington fulgurite was sand and basalt gravel berm with oxide compositions listed in Table 1 and may have had a portion of aluminium conductor melted into the fulgurite. We believe, in contrast, that the Southwick fulgurite is fully natural while the Farmington fulgurite is a mix of natural and anthropogenic target materials. We highlight the Farmington fulgurite here mainly to show that the similarities between the Southwick and Farmington bulk compositions shows that these fulgurites were formed from similar materials. As samples of the surrounding soil were not available for confirmation of the Southwick fulgurite's formation site, such a comparison helps verify that the fulgurite was indeed found in its stated region.

Samples were analysed using a Zeiss-EVO MA15 Scanning Electron Microscope (SEM) with attached Oxford INCA250 Energy-Dispersive Spectrometer (EDS). The operating parameters were a probe current of 500 pA, counting time of 500 s, average count rate for the whole spectrum of 2500 cps, and operating accelerating potential of 25 kV. A 30 nm carbon film was sputter-coated onto the samples.

The Fe–Si phases were analysed with a Bruker D8 Venture equipped with a Photon II CCD detector single-crystal X-ray Diffractometer (XRD). The instrument utilised graphite-monochromatised $\text{MoK}\alpha$ radiation ($\lambda = 0.71073 \text{ \AA}$), a detector-to-sample distance of 7 cm, and an exposure time of 30 s per frame. Intensity data was integrated for adsorption and Lorentz-Polarization with software packages from Bruker. Both

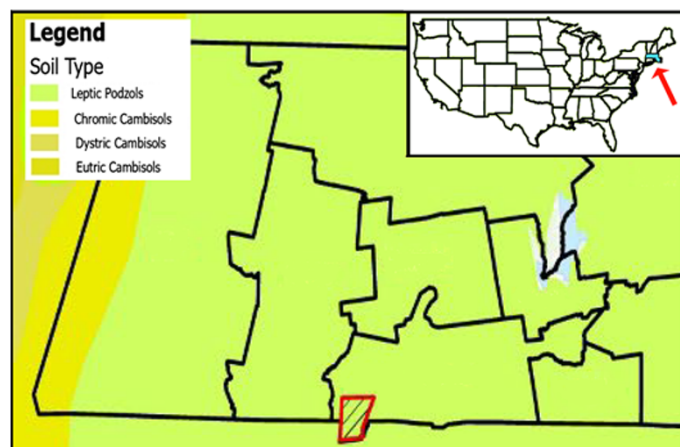


Figure 2. Map of Massachusetts (located in blue on the inset map of the USA) with Southwick highlighted in red. The area of Southwick is comprised of leptic podzols which are typically found within 200 cm from the surface and comprised of appreciable clay mineralogy (Kitagawa, 2005).

Table 1. Oxide weight percentages of Farmington Connecticut fulgurite as per Sheffer (2007), for contrast with the Southwick Massachusetts fulgurite in Table 2

	Bulk glass	SiO ₂	'Lower', homogenous glass
SiO ₂	55.83	98.85	76.35
FeO	6.32	0	4.03
TiO ₂	19.92	0	0.48
Al ₂ O ₃	2.91	0	10.35
MgO	6.00	0	1.45
K ₂ O	1.48	0	1.35
CaO	5.09	0	2.46
Na ₂ O	3.53	0	0.80
Total	101.08	98.85	97.27

XRD and SEM were performed at the CRIST, Centro di Studi per la Cristallografia Strutturale, Department of Chemistry, Università di Firenze, Italy

We used thermodynamic data to investigate the formation of iron silicides within the Southwick fulgurite. Specifically, the Gibbs free energy (ΔG) and equilibrium constant (K_{eq}) of three potential reactions investigating the transformation of one iron silicide into another were calculated using a range of temperatures from 298.15 – 1773.15 K increasing at increments of 25 K. The specific heat capacity (c_p), ΔH (change in enthalpy) and ΔS (change in entropy) of Fe₅Si₃ were calculated using *HSC chemistry workbench* (a thermodynamic equilibrium modelling program from Outokumpu Research Oy; for more details see Pasek and Greenberg, 2012). S^0 , ΔH and c_p values at 298.15 K for SiO₂, O₂, Fe, and Si were obtained from the NIST chemistry webbook (Linstrom, 1998). For all subsequent values past 298.15 K, a midpoint formula was used where thermodynamic values of the enthalpy (H) and entropy (S) were estimated at the 25 K incremental steps.

$$H_T^0 = \Delta H_{298.15}^0 + \int_{298.15}^T c_p dT \quad (2)$$

$$S_T^0 = \Delta S_{298.15}^0 + \int_{298.15}^T \frac{c_p}{T} dT \quad (3)$$

Using the c_p of Fe₅Si₃ the c_p for Fe₂Si was calculated by multiplying the c_p Fe₅Si₃ by 3/8ths as c_p follows the number of atoms for related species. The Δc_p of Fe₂Si was calculated by obtaining a c_p value by taking an average of c_p at 298.15 K gathered from Cui and Jung (2017) for Fe₅Si₃ (Cui and Jung, 2017) and subtracting the summation of iron and silicon c_p and dividing the result by 2. The ΔH value for temperatures past 298.15 K for Fe₂Si was calculated using equation 2. The ΔH of Fe₂Si at 298.15 K was obtained through an averaging of values obtained from Cui and Jung (2017).

Using equation 3, the S^0 for Fe₂Si were calculated for subsequent temperature past 298.15 K. ΔS of Fe₂Si was calculated using the average S^0 value at 298.15 K obtained from Cui and Jung (2017) and subtracting the summation of iron and silicon S^0 values. The ΔG and K_{eq} were then calculated using subsequent equations, below.

Results

The Southwick Massachusetts fulgurite consists primarily of a groundmass of oxide glass (65%) with lechatelierite (200–400 μ m in diameter, compositionally >99% SiO₂) dispersed throughout (35%). Accessory phases include spinel and metal grains. The groundmass of the fulgurite was analysed by SEM to constrain

Table 2. Oxide weight percentage of Southwick Massachusetts fulgurite groundmass glass

Oxide	Weight %
SiO ₂	61.06
FeO	7.73
TiO ₂	1.16
Al ₂ O ₃	15.11
MgO	3.43
K ₂ O	1.50
CaO	6.76
Na ₂ O	3.26

its composition (Table 2) and investigate any unusual minerals within the glassy matrix. The general composition of the fulgurite seems to match with the information gathered from the Southwick quadrangle, but the composition is like the nearby Farmington fulgurite, specifically the 'lower' glass, albeit with more Al₂O₃ in the Southwick fulgurite (Table 1). The predominant composition of the protolith of the Southwick fulgurite is a clayey till which tracks with higher Al₂O₃ percentages in contrast to the Farmington fulgurite. The two fulgurites are compositionally similar if the SiO₂ of the Farmington fulgurite is reduced from 75 to 61 wt.%, and all other oxides are increased by 50% by weight (Supplementary Figure S1).

Notably, in general the composition of the protolith for most fulgurites closely tracks the composition of the outer glass (a 'baked' material), especially for Type II fulgurites (see description below), as the temperature of this 'baked' zone does not exceed the temperature of vaporisation of the major rock-forming elements. However volatiles such as organic carbon and water are both depleted during the fulgurite-forming process (e.g. the Winans lake fulgurite, despite forming along a tree root, contained only trace quantities of graphite, see Essene and Fisher, 1986).

One spherical grain (~95 μ m in diameter) was found to exhibit a reaction rim (Fig. 3) with an outer rim of Fe₅Si₃ and a core of Fe₂Si. This was the only metal spherical grain noted within this section of the fulgurite. Based on SEM EDS data of this grain, the outer rim has an iron atomic percentage of ~62% whereas the core is close to 67%. For a correct identification, the two phases were handpicked from the polished section and analysed using single-crystal XRD. The outer rim turned out to exhibit a hexagonal symmetry, with $a = 6.76(2)$ Å and $c = 4.72(2)$ Å, closely resembling the mineral xifengite, Fe₅Si₃. In contrast, the core corresponds to the mineral hapkeite, Fe₂Si, with cubic symmetry and cell value of $a = 2.85$ Å.

Discussion

The iron silicide phases seen in the Southwick Massachusetts fulgurite include an external rind of Fe₅Si₃ and an internal core of Fe₂Si. This section will discuss the potential routes of formation of iron silicides as well as the potential significance of this grain. Following the fulgurite classification system, this fulgurite best fits the Type II archetype of clayey soil being struck by lightning (Pasek et al., 2012). Iron silicide in fulgurites generally fall into two groups: iron rich and silicon rich, and most fulgurites bear silicon-rich silicides (Si > Fe by atomic fraction), if any are even present. Iron-rich silicides are more common in extraterrestrial material (Feng et al., 2021), but do occur in fulgurites. The fulgurite from Southwick, Massachusetts, falls within the iron-rich category, with iron metal

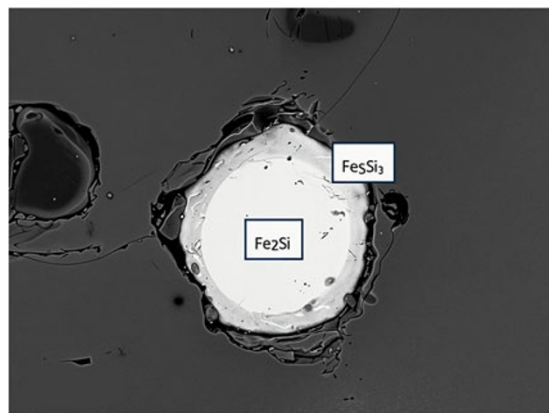


Figure 3. Back-scattered electron image of grain of Southwick Massachusetts fulgurite exhibiting a reaction rim. The outer rim is made up of Fe_5Si_3 and the core is Fe_2Si . Sphere diameter is 95 μm , including rim.

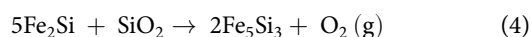
Table 3. Selected ΔG values for equations 4, 5 and 6

Temperature (K)	ΔG equation 4	ΔG equation 5	ΔG equation 6
298.15	1852.8	-116.84	-31.753
673.15	1407.8	-280.52	-166.22
1048.15	896.41	-591.56	-405.87
1423.15	219.03	-974.28	-698.47
1773.15	265.30	-1570.1	-1152.2

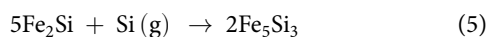
potentially reacting with all reduced silicon prior to the formation of more Si-rich silicides or even elemental silicon.

To further understand the mechanisms behind this reaction rim the thermodynamic data (ΔG in kJ/mol) for three possible equations were calculated (Table 3, with full range of values available in Supplementary material). Note that for these calculations we assume a pressure of 1 atm, and since some fulgurites may experience high pressure conditions, that is a limitation of the current model. We also acknowledge that applying thermodynamics to a system that may not have time to reach equilibrium necessarily means that the results predicted from thermodynamics are at best a prediction of reaction direction, and not a direct reflection of reality.

We tested three potential routes for the formation of a Fe_5Si_3 rind from Fe_2Si to see which may be the most plausible. In the first reaction, the internal Fe_2Si core may be reacting with SiO_2 melt (either as SiO_2 melt that later formed lechatelierite that comprises 35% of the fulgurite by volume, or as a component of the groundmass melt), and expelling O_2 to the environment.



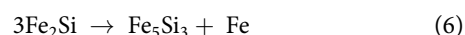
The second potential route of formation would follow the reaction:



In this reaction, the Fe_2Si core is reacting with silicon gas, through a silicidation reaction. This would parallel the known sulfidation of metals in meteorites (Imae, 1994) and the phosphidation of metals, for example within Lunar impact rocks (Gooley *et al.*, 1973; Pasek, 2015). Notably, this reaction requires the presence of $\text{Si}(\text{g})$, which must have originated from reduction of the soil (e.g. as per Essene and Fisher, 1986). In addition, equation 5 is similar to equation 4, in as much as SiO_2 may break down to Si and O_2 gases, through a thermodynamically unfavourable reaction. However, in

the presence of organic carbon, the generation of CO and CO_2 gas may push the reaction towards products.

The third potential route of formation would be by incongruent melting, but this seems to be the least likely option as a more reduced phase comes from an oxidised phase, and is inconsistent texturally with the sample. That said, such a disproportionation reaction is expected from some reducing environments, such as the formation of awaruite (Bird and Weathers, 1975, 1979). Incongruent melting of minerals within fulgurites has also been reported for zircons (Kenny and Pasek, 2021).



The rind we see is more appropriately termed a reaction rim. A reaction rim signifies a secondary mineral that forms a rim around a primary grain. Possible causes of formation of these reaction rims are: (1) early formed crystal reactions with a melt; (2) solid-state reactions with an intergranular fluid such as gas; (3) magmatic accretion from a different liquid; and (4) solid-state reactions between grains adjacent to primary during thermal or regional metamorphism (Spry, 2013).

Reaction rims can be found as a simple single layer surrounding a grain. A prime example of this is the serpentinisation process where serpentine minerals form through water/rock interaction and begin encrusting olivine grains in peridotite (Cuthbert, 1990). Reaction rims can also form in a complex process with an assemblage of minerals. For example, one clearly defined rim from Griffin and Heier (1973) showed an olivine grain (rimmed by accessory minerals such as orthopyroxene, clinopyroxene, garnet and spinel) next to a plagioclase grain. Following the initial growth structure, garnet began to propagate outwards and enveloped the previous rims (with previous grains becoming inclusions) with clinopyroxene forming. The final stages of this reaction rim combine the reactions of water, garnet and clinopyroxene to form a hornblende layer.

Seemingly, the closest mechanism that describes this fulgurite's reaction rim is a reaction between adjacent grains during thermal metamorphism. The thermal aspect could be attributed to the intense heat from the lightning strike. As such, this lightning-induced melting may be similar to prior reports of lightning-struck rocks (Castro *et al.*, 2020). It may also be possible that the lightning strike created a reducing environment further promoting the reduction from core to rim species. Based on thermodynamic data, the most likely reaction is the silicidation reaction in equation 5. This reaction requires a very low f_{O_2} (Essene and Fisher, 1986 estimate an f_{O_2} between 10^{-18} to 10^{-13} at 1700 K).

We favour equation 5, in which silicon gas reacts with iron silicides to generate new iron silicides. Equation 5 is an exergonic reaction (Table 3), and we also expect increasing silicidation with higher pressure as temperatures increase (which results in a favouring of the products because gas is a reactant in equation 5, vs. a product in equation 4). Pressure may also increase due to shock (Chen *et al.*, 2017). We base this on the Fe-Si phase diagram for these phases (Fig. 4). From this figure it is apparent that Fe_2Si is a higher temperature phase than Fe_5Si_3 , with Fe_2Si being stable from 1317–1476 K, whereas Fe_5Si_3 is stable from 1093–1364 K. Given that the other major occurrence of rinds due to incongruent melting in fulgurites is on zircon with rinds of baddeleyite ($\text{ZrSiO}_4 = \text{ZrO}_2 + \text{SiO}_2$) (e.g. Pasek *et al.*, 2012; Kenny and Pasek, 2021), these rinds form because of the high-temperature phase forming a rind on the low-temperature phase. The occurrence of Fe_5Si_3 as a rind on Fe_2Si is the opposite.

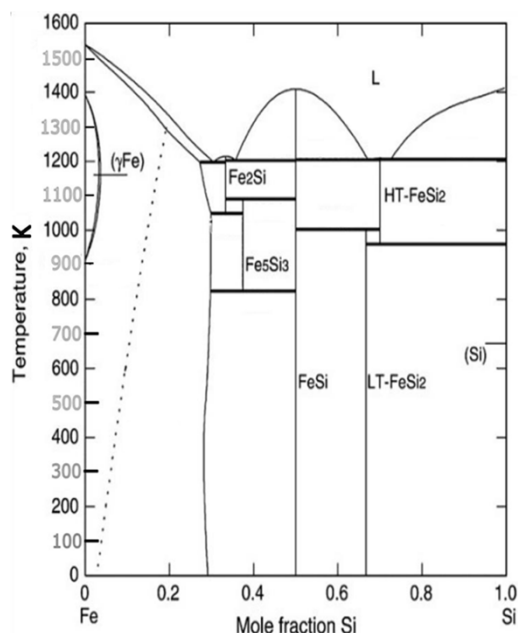


Figure 4. Calculated Fe–Si phase diagram according to the modelling by Lacaze and Sundman (Lacaze and Sundman, 1991) and modified from Liang *et al.* (2011).

As a ‘sanity check’ we contrast the sphere in Fig. 3 to known metal gas–solid kinetics (e.g. oxidation, see Supplementary material). Metals generally follow a parabolic rate law during gas–solid reactions (Monceau and Pieraggi, 1998), where the mass gained per surface area is proportional to the square root of the reaction time. If we assume a reaction time of one second, then for a sphere with a 100 μm diameter, we find a requirement of a parabolic rate constant of $\sim 5 \times 10^{-6} \text{ kg}^2/\text{m}^4\text{s}$. This rate constant matches measurements of the parabolic rate constant for iron phosphidation (Sasaki, 1985) at high temperature and, probably, under high pressure (see Supplementary Material), as unfortunately metal silicidation rates have not been measured as robustly. However, the silicidation of metal should be reasonable under these timescales.

Conclusions

Iron silicides have been known from fulgurites since their discovery by Essene and Fisher (1986) in the Winans Lake fulgurite from Michigan, USA. These materials clearly indicate highly reducing conditions during the formation of a fulgurite from a lightning strike, usually from combusting organic matter. The likely driver of the reduction in the Southwick fulgurite is similarly combusting organic matter, which drives the oxidation state to extreme reduction, though no organic material was found within the Southwick fulgurite. Organic material has however been observed in other fulgurites, in the form of graphite (Essene and Fisher, 1986), fullerenes (Daly *et al.*, 1993), polycyclic aromatic hydrocarbons (Carter *et al.*, 2010; Elmi *et al.*, 2017) and partially graphitised carbon (Feng *et al.*, 2019). It seems likely that a similar organic driver promoted reduction of the matrix to Fe_2Si and Fe_5Si_3 in the Southwick fulgurite.

The fortuitous discovery of a Fe_5Si_3 rind on an Fe_2Si core within the Southwick fulgurite may give clues as to how these minerals are generated during a lightning strike. We note that, as only one grain was observed within this fulgurite, silicides may form

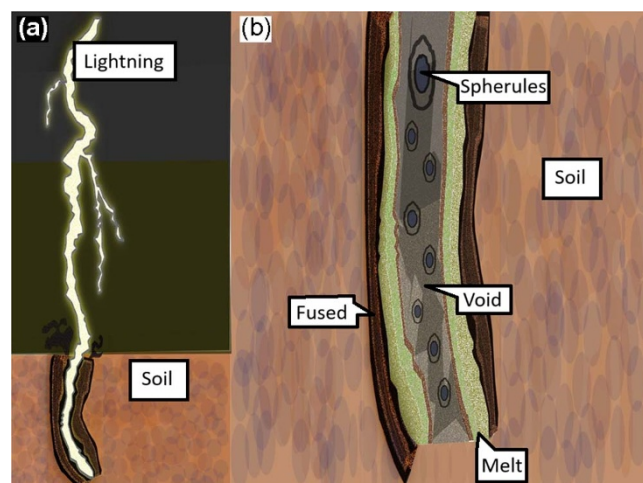


Figure 5. An illustration showing: (a) lightning striking and propagating through soil; the lightning vaporizes a portion of the soil leaving a void space and Si gas; (b) as this gas cools, iron-metal rich grains, possibly as Fe_2Si solidify or are formed as liquid droplets, these droplets then react with Si in the gas, forming the Fe_5Si_3 rind around the Fe_2Si ; after a few seconds the fulgurite cools and a majority of the gas condenses as a glass, entraining the silicides within the glassy matrix.

by an assortment of other processes, however for this grain, we propose that the most likely pathway for this grain’s formation is through silicidation of existing iron silicides. This sphere probably formed through gas–solid reactions in the cooling lightning plasma channel. Silicides are unstable at the peak temperatures of lightning forming a fulgurite ($>1800\text{K}$), and hence they had to form as the fulgurite cools. The initial Fe_2Si phase probably formed under extremely reduced conditions somewhere between the iron–wüstite and the silicon–silica buffers, and probably favoured this composition due to the lower SiO_2 abundance of the protolith. For example, Feng *et al.* (2021) track Fe–Si composition in fulgurites compared to groundmass composition, finding that fulgurites with ~ 60 wt.% or less SiO_2 tend to have more Fe-enriched silicides (where $\text{Fe} > \text{Si}$ by atomic fraction), whereas those with $>70\%$ SiO_2 tend to have Si-enriched silicides.

We present a simple illustration of the likely formation process for these grains (Fig. 5). In this process, lightning strikes and propagates through soil (Fig. 5a), vaporising a portion of the soil. As this gas cools, grains rich in iron metal, possibly solidify as Fe_2Si or are formed as liquid droplets (Fig. 5b). These droplets then react with Si in the gas, forming the Fe_5Si_3 rind around the Fe_2Si . After a few seconds the fulgurite cools and the gas condenses as a glass, entraining the silicides within the glassy matrix. This may also suggest that, as the fulgurite cools, the gas may have become more reducing, given that the silicon content of the grain increased in the formation of the reaction rim. This may have been due to separation of oxygen from the environment, perhaps as an expelling of CO_2 .

This reaction sequence may explain at least some of the silicides found in fulgurites (but probably not all). Iron silicides in fulgurites can form by reaction of gas with a condensed phase (most likely solid iron metal), and the Southwick fulgurite seems to be capturing one grain in the process of this reaction. Notably, gas–solid reactions can be extremely fast, and similar rinds ($\sim 10 \mu\text{m}$ in diameter) have been found in lunar impact rocks, suggesting a timescale of seconds, and reaction kinetics—at least for P reacting

with Fe—would reasonably form rinds of the size observed in the Southwick fulgurite.

Supplementary material. The supplementary material for this article can be found at <https://doi.org/10.1180/mgm.2024.108>.

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Competing interests. The authors declare no competing interests.

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