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Chapter 23

Non-magmatic glasses

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Overview

On Earth, natural glasses are typically produced by rapid cooling of melts, and as in the case of minerals and rocks, natural glasses can provide key information on the evolution of the Earth. However, natural glasses are products not solely terrestrial and different formation mechanisms give rise to a variety of natural amorphous materials. In this chapter, we provide an overview of the different natural glasses of non-magmatic origin, and on their formation mechanisms. Here we focus on natural glasses formed by mechanisms other than magmatic activity. Included are metamorphic and glasses from highly energetic events (shock metamorphism). The study of these materials has strong repercussions on planetary surface processes, paleogeography/paleoecology and even on the origin of life.

Outline:

Introduction

1. The composition and origin of natural glasses

- Brief description of the most common natural amorphous materials and of the different mechanisms of formation.

2 Metamorphism

2.1 Pyrometamorphic glasses

2.2 Pseudotachylites

3. Glasses from highly energetic events (shock metamorphism)

3.1 - Impactites

3.1.1 – Distal ejecta

Tektite and microtektites

Enigmatic Impact glasses

3.1.2 – Proximal impactite - suevite

3.2 fulgurite

Case study: fulgurite.

3.3 trinitite

4. Glass properties

43 **Acronyms & Glossary**

- 44 Hypervelocity impacts = Impacts, involving impacting bodies that are traveling at speed
45 (generally greater than a few km/s) higher enough to generate shock waves upon impact.
46 K-Pg = Cretaceous–Paleogene (K–Pg) boundary (~66 million years ago)
47 KT = Cretaceous-Tertiary boundary (former name for K-Pg)
48 Lechatelierite = shock-fused SiO₂ glass
49 LDG = Libyan Desert Glass
50 m-T = microtektites; Small distal ejecta with diameter less than 1 mm.
51 MN = Muong Nong-Type tektites
52 PDFs = Planar deformation features; microscopic parallel, isotropic features in minerals that
53 originate from elevated shock metamorphism.
54 SF = tektite/impact glass strewn fields
55 TAS = Total-Alkalis versus Silica diagram
56 Tektites = millimeter- to centimeter-scale, glassy particles of ballistically transported impact
57 melt, formed by the impact of an extraterrestrial projectile.
58 T_g = Glass Transition temperature
59 YD = Younger Dryas is a geological period from ~ 12,900 to ~ 11,700 BP
60 YDB = Younger Dryas boundary
61 XAS = X-ray Absorption Spectroscopy
62

INTRODUCTION

63
64

65 Natural glasses have accompanied human civilization and have always aroused human
66 curiosity. Overtime, mankind tried, first of all, to understand the origin and the properties of
67 these materials, and then, they have tried to recreate and improve, the natural occurring
68 specimens. That is what brought us into the “Glass Age”, and nowadays, we cannot think of a
69 life without glasses. Indeed, from rudimental weapons, decorative objects, or storage bottles,
70 currently glasses are more high-tech materials used for example for high-speed
71 communications, or in smart-windows. Many smart materials have been developed from the
72 observations done on natural ones, but besides the material technology, are the natural sciences
73 (*s.l.* incorporating life and physical sciences) that have gained the highest contribution from the
74 study of natural amorphous materials. They have been used to explain why and when natural
75 phenomena occurred, and to obtain a general understanding of natural phenomena. For
76 example, in geosciences, knowledge about past events (geologically speaking) either on Earth
77 or other terrestrial planets is often derived by the study of magmatic and non-magmatic glasses.
78 Besides geosciences and engineering, the study of natural aluminosilicate glasses has been a
79 key topic in the framework of waste managements, especially for evaluation of the long-term
80 stability of nuclear waste form glasses (Chapman *et al.* 1984; Ewing 1979). Natural glasses
81 may remain stable for very long periods (more than 1 Ma) in various geological settings and
82 climatic conditions, in contrast to many manmade glasses from the Middle Age. Hence, natural
83 glasses are used as analogues for extrapolation of short-term laboratory data (hours to years) to
84 the long time periods (hundred to thousand years).

85

86 The aim of this chapter is to review natural glasses formed by mechanisms other than
87 magmatic activity, and after a brief description of the most common natural amorphous
88 materials and of the different mechanisms of formation, we report some examples on

89 metamorphic glasses, and on glasses from highly energetic events. At the end we report an
90 overview of the properties of these natural amorphous materials.

91 In the last decades, several reviews, for specific natural glasses, have been published and
92 the readers are referred to these exhaustive papers [*e.g.* [Eby *et al.* 2010](#); [Glass 1990](#); [Glass &
93 Simonson 2013](#); [Koeberl 1997](#); [Pasek *et al.* 2012](#)]. More recent general reviews on different
94 natural glasses are provided in [Cicconi & Neuville 2019](#); [Heide & Heide 2011](#); [McCloy 2019](#).

95

96 **1. The composition and origin of natural glasses**

97

98 Natural glasses have different origins and chemical compositions, though, their chemical
99 variability match the differentiations found in many common types of volcanic rocks. An
100 overview of the enormous variability of natural glasses occurring on Earth and on Lunar soil
101 can be appreciated in the TAS diagram (total alkali vs. silica; [Figure 1](#)). These amorphous
102 materials have SiO₂ contents between ~33-99 wt.%, and alkali contents ranging between 0 and
103 15 wt.%, thus covering many common types of volcanic rocks. Despite the fact that these
104 glasses are differentiated chemically as many common volcanic rocks, most of the times, non-
105 magmatic glasses experienced extreme conditions of formations, far away from those of
106 common igneous materials. Indeed, most of the glasses considered in this chapter have cooling
107 rates orders of magnitude higher than volcanic terrestrial (or lunar) glasses, and/or have been
108 subject to extremely elevated peak pressures. For instance, cooling rates may range from
109 extremely high values, as in fulgurite formation (~10¹⁰ °C/min) and submarine basaltic
110 eruptions (10⁷ °/min), to moderate values in tektites 10⁴ -10⁻² °/min, to very slow rates in
111 massive obsidian flows 10⁻² – 10⁻⁴ °/min ([Potuzak *et al.* 2008](#); [Rietmeijer *et al.* 1999](#); [Switzer
112 & Melson 1972](#); [Weeks *et al.* 1984](#); [Wilding *et al.* 1996b, 1996a](#)). Out of curiosity, water, which

113 is considered to be a very weak glass former, requires cooling rates faster than $10E6$ °/min to
114 form an amorphous solid.

115

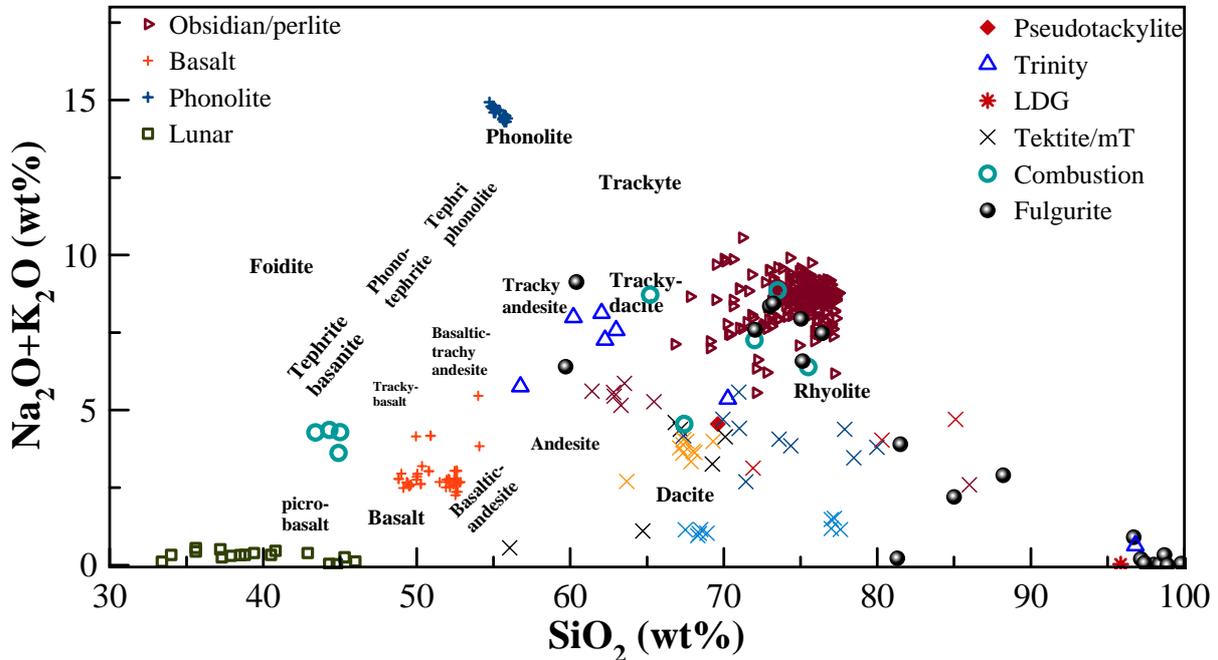
116 Before describing non-magmatic glasses, we would like to provide an overview of all
117 amorphous materials found on Earth (and on other terrestrial planets), and of the various
118 mechanisms of formation. The most known natural amorphous materials are of magmatic origin
119 and include basaltic glasses and obsidians. Basaltic glasses have an average composition of
120 about (wt.%) 50-54% SiO_2 , 12-17% Al_2O_3 , 8-12% FeO_{tot} , 2-4% alkali ($\text{K}_2\text{O} + \text{Na}_2\text{O}$), 15-20%
121 alkali-earth ($\text{CaO} + \text{MgO}$) (Cicconi & Neuville 2019 and references therein) and their low
122 viscosity favors crystallization (devitrification). Volcanic glasses produced upon rapid cooling
123 of basaltic melts are called sideromelane, but they also occur as volcanic ash, fibers and
124 teardrops (*i.e.* Pele's Hair and Pele's Tears) and more rarely form solidified foam – reticulite
125 (Cicconi & Neuville 2019). Obsidian glass was first described by Pliny The Elder's Natural
126 History with the name of “*obsiana*”, so called because its similarity to a very dark stone found
127 in Ethiopia by Obsius. This glass has accompanied and influenced human evolution since
128 prehistoric times, and nowadays, it still enters the popular culture, even if with more fanciful
129 name (*i.e.* dragonglass, after the fantasy drama television series Game of Thrones). Obsidians
130 are generally subalkalic rhyolitic with an average composition of about (wt.%) 72-77% SiO_2 ,
131 10-15% Al_2O_3 , 1-2% FeO_{tot} , 7-10% alkali ($\text{K}_2\text{O} + \text{Na}_2\text{O}$), 0.5-2% alkali-earth ($\text{CaO} + \text{MgO}$)
132 (Cicconi & Neuville 2019 and references therein). The glass-forming processes of obsidian
133 melts are strongly influenced by the content, size and shape of microlites (Castro *et al.* 2002),
134 and by the contents of volatile components (such as water, fluorine and chlorine, sulphur and
135 carbon oxides (Carmichael 1979; Heide & Heide 2011), and small variations in volatile
136 contents can cause important changes in the flow dynamics of obsidian melts.

137 Other natural amorphous materials are formed by metamorphic processes. For instance,
138 the so-called pyrometamorphic glasses form due to burning of fossil fuels such as coal and
139 natural gas or other organic material (McCloy 2019 and references therein). The term
140 pyrometamorphism, which defines a type of contact metamorphism, was originally proposed
141 in 1912 by Brauns (Grapes 2010 and references therein) to describe a high-temperature/ low
142 pressure metamorphism observed in schist xenoliths in trachyte and phonolite magma of the
143 East Eifel area (Germany). The term buchite is used to define those partially or completely
144 melted materials as a consequence of pyrometamorphism.

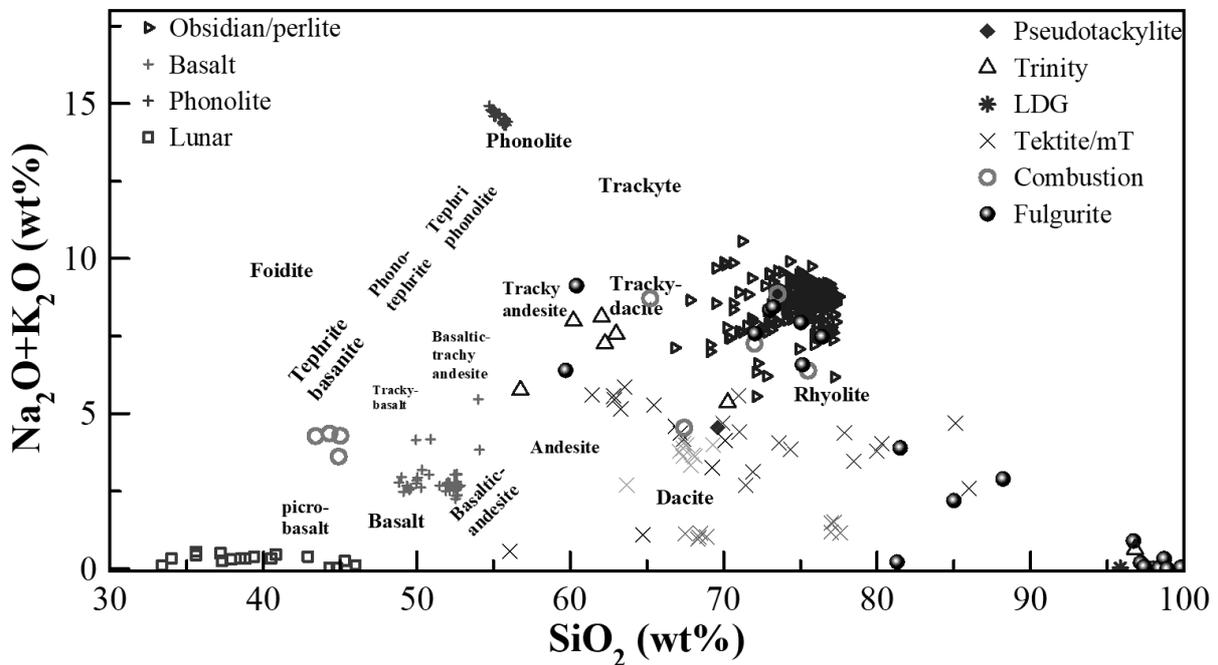
145 Glasses from highly energetic events are formed in a completely different way, and with
146 a completely different timescale than magmatic and metamorphic ones. For instance, impact
147 glasses formation is related to the collision of an extraterrestrial body on the surface of the
148 Earth. Thus, they derive from shock metamorphism of existing silicate rocks and sediments.
149 Impacts or airbursts (shock melting caused by a cosmic object exploding in the atmosphere)
150 can be either natural or artificial, and both provide shock markers due to the extreme high
151 temperatures and pressures. Among natural glasses that experienced extremely high
152 temperatures in a very short time there are also the fulgurites, formed following lightning strike.

153 Finally, biomineralization processes can produce amorphous materials. These are
154 considered eco-friendly and thus they have captured the attention either of organic-/inorganic-
155 chemists or materials scientists. Several siliceous marine organisms exhibit discontinuous,
156 three-dimensional frameworks of short chains of SiO_4 tetrahedra, bonded with apical
157 hydroxyls. The low-temperature hydrated variety of silica, opal ($\text{SiO}_2 \cdot n\text{H}_2\text{O}$), is a biomineral,
158 and displays different arrangements of amorphous SiO_2 , water, and cristobalite, and/or
159 tridymite (Cicconi & Neuville 2019). Depending on the arrangements, it is possible to
160 distinguish three opal types: i) opal-C (cristobalite); ii) opal-CT (cristobalite and tridymite); iii)
161 opal-A (X-ray-amorphous opal). The latter can be further divided in opal-AN (*e.g.*, hyalite) and

162 opal-AG with an amorphous silica gel structure. In a “maturation process” (Ostwald ripening)
 163 opals are transformed as follows (Skinner & Jahren 2003 and references therein): Opal-AG →
 164 Opal-CT → Opal-C → microcrystalline quartz.



165



166

167 **Figure 1** – SiO₂ vs. total alkali (wt%) diagram for several natural non-magmatic glasses
 168 described in this chapter. Glasses deriving from magmatic processes are reported for
 169 comparison. Data compilation from [Cicconi & Neuville 2019](#). LDG = Libyan Desert Glass;
 170 mT = microtektites.

171

172

173 **2. Metamorphic glasses**

174

175 **2.1 Pyrometamorphic glasses**

176 2.1.1 General features of pyrometamorphism

177 Pyrometamorphic glasses are in important class of amorphous materials summarized by
178 (Grapes 2010). The term ‘pyrometamorphism’ was originally used by (Brauns 1912) to describe
179 changes in contact zones (*i.e.*, aureoles) between magma intrusions and country rock, where
180 high temperature and atmospheric pressure result in particular mineralogical and morphological
181 changes. These high temperature changes are often observed in xenoliths within later igneous
182 rocks, near shallow magmatic intrusions, and within tuffs and breccias. Brauns assumed that
183 pyrometamorphism must create melting in the heat-affected rocks and vitrification on cooling.
184 Other terms have been introduced but have fallen out of favor, such as optalic metamorphism,
185 emphasizing the transient, quickly dissipated heat such as used when ‘baking a brick.’
186 Pyrometamorphized rocks tend to lose all volatiles, and melts recrystallize with anhydrous
187 mineral assemblages. Some characteristic evidences of such processes include bleaching of
188 carbonate rocks, reddening of iron-containing rocks, fusion of mineral grains, hardening, and
189 similar morphological and chemical changes similar to those observed in earthenware ceramic
190 and clay brick-making processes. Heat for pyrometamorphic transformations need not come
191 from contact magmatic heat, but also from other sources, such as combustion of coal beds and
192 organic-rich sediments or even lightning. The Subcommittee on the Systematics of
193 Metamorphic Rocks (SCMR) of the International Union of Geological Sciences (IUGS)
194 categorizes these effects as subvariants of contact metamorphism, namely ‘burning/combustion
195 metamorphism’ and ‘lightning metamorphism,’ *i.e.*, fulgurites (Callegari & Pertsev 2007).

196 While contact and lightning metamorphism are highly localized, high temperature
197 changes in rock due to coal bedding combustion can be regionally extensive. Well documented
198 examples in the USA occur in the Grimes Canyon area, Monterey Formation, California

199 (Bentor *et al.* 1981); the Powder River Basin, Wyoming (Cosca *et al.* 1989; Herring &
200 Modreski 1986); and various coal-bearing areas of Montana and Colorado (Rogers 1918). The
201 phenomenon of combustion metamorphism has been found to be quite common (Bentor *et al.*
202 1981; Grapes 2010), including examples in China (de Boer *et al.* 2001), Mongolia (Peretyazhko
203 *et al.* 2018), India, Russia (*e.g.*, Chelyabinsk brown coal basin) (Sokol *et al.* 1998), Iran, Jordan
204 (Khoury *et al.* 2015), Israel (Ron & Kolodny 1992; Sokol *et al.* 2014), Czechia, Germany,
205 England, Italy (Melluso *et al.* 2004), Mali (Svensen *et al.* 2003), Canada (Canil *et al.* 2018;
206 Mathews & Bustin 1984), New Zealand, and Australia.

207 Some terms are commonly used for pyrometamorphic rocks as follows. Hornfels forms
208 where clay-rich rocks contact a hot igneous body, and partially or completely recrystallize after
209 in-situ melting, resulting in baked and hardened silicate + oxide systems. Clinker originally
210 referred to altered or burned coal, but now refers generally to burnt brick-like rock which ‘rings’
211 (‘clinks’) when struck. Note that clinker is also the high-temperature processed calcium
212 aluminosilicate used as the reactive precursor for Portland cement. These same ‘clinker’
213 minerals, *e.g.*, belite sulfoaluminate, have been found in Israel in natural pyrometamorphic
214 contexts (Sokol *et al.* 2014). Buchite, in contrast to hornfels, is largely vitrified from high heat
215 applied to sandstones or pelites, resulting in hard, fused, and glassy material; the term is not
216 restricted to coal fire lithologies, so in that case should be referred to as ‘buchite clinker.’
217 Finally, paralava resembles artificial slag or basalt, is generally vesicular and aphanitic,
218 sometimes shows evidence of flow, and formed from burning and melting of sedimentary rocks
219 (shale, sandstone, marl) by proximal combusting coal seams. The distinction among these is
220 based on both protolith and peak temperature, which can be ~ 400 to >1600 °C locally, resulting
221 in a continuum from baked/burnt rock to fused-grained rock (clinker) to partially melted
222 buchite, to wholly melted and partially devitrified rock (paralava), often all in the same area.
223 Terminology is not uniform employed, and in certain localities clinker is still be called ‘scoria’

224 despite the different volcanic origin assumed from that term. Use of the terms ‘buchite’ and
225 ‘paralava’ imply the observable presence of glass resulting from a quenched rock melt ([Grapes](#)
226 [2010](#)).

227

228 2.1.2 Coal fires and spontaneous combustion

229 Coals in the Western USA, known to spontaneous combust, are typically lignite or sub-
230 bituminous grade (Rogers, 1917). Spontaneous combustion occurs due to a complex process
231 involving absorption of oxygen, such as through cracks, and oxidation of unsaturated
232 hydrocarbons. The main exothermic reaction is that of carbon reacting with oxygen gas to form
233 CO₂ ([Grapes 2010](#)). These early reactions take place at temperatures as low as 80 °C, but this
234 oxidation further generates heat until about 200 °C, where autogenous oxidation occurs,
235 followed by ignition at 350-400 °C. Fine dust lignite, however, which has typically absorbed a
236 large amount of oxygen, can ignite at temperatures as low as 150 °C ([Rogers 1918](#)). Moisture
237 is said to exacerbate this effect, with escaping hydrogen-containing gases also playing a role.
238 Other factors influencing the combustion reaction include coal factors such as rank and pyrite
239 content, reaction factors such as particle size and temperature, and macroscopic factors such as
240 air flow, overbedding rock type, and thermal conduction ([Grapes 2010](#)). Often areas can burn
241 in the absence of additional oxygen, producing highly reducing conditions and the creation of
242 coal ash with evidence of metallic iron ([de Boer et al. 2001](#); [Grapes 2010](#)).

243 Combustion of coal beds has been observed in modern times on the coast of Dorset,
244 England; in the ‘Smoking Hills’ of Canada ([Mathews & Bustin 1984](#)); and at the ‘Burning
245 Mountain’ (Mt. Wingen) in New South Wales, Australia ([Bentor et al. 1981](#)). The Smoking
246 Hills are thought to have been burning for at least 150 years, while the Burning Mountain has
247 been combusting for at least 15,000 years. The Posidonia shales in Germany, though not

248 currently combusting, are thought to have been active in the Middle Ages (Bentor *et al.* 1981).
249 Many other examples are known (Kuenzer & Stracher 2012).

250 Examples of modern creation of pyrometamorphic materials are known from burning of
251 a wide range of fossil fuels, from spontaneous combustion but also other natural or
252 anthropogenic ignition (Kuenzer & Stracher 2012). Combustion in coal mining ‘spoil heaps’
253 has resulted in recent pyrometamorphic processes in Russia (Chelyabinsk) (Sokol *et al.* 1998)
254 and Italy (Ricetto) (Capitanio *et al.* 2004; Stoppa *et al.* 2005) (Figure 2). One of the interesting
255 features of the assemblage in Russia is the presence of pure glassy carbon, known as shungite.
256 Additionally, minerals have been observed which were crystallized from the gas phase in
257 cracks, including pure oxide, sulfide, silicate, and carbide single crystals.

258

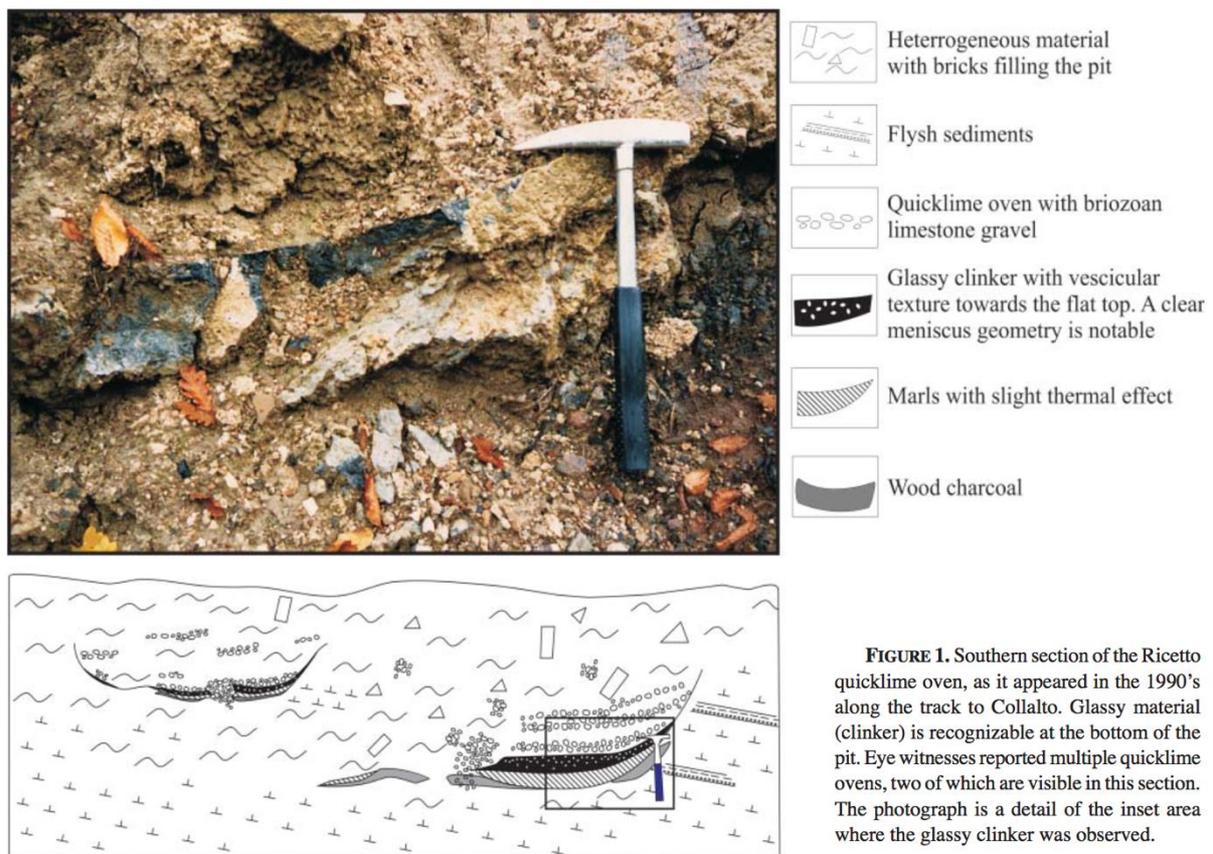


FIGURE 1. Southern section of the Ricetto quicklime oven, as it appeared in the 1990's along the track to Collalto. Glassy material (clinker) is recognizable at the bottom of the pit. Eye witnesses reported multiple quicklime ovens, two of which are visible in this section. The photograph is a detail of the inset area where the glassy clinker was observed.

259

260 **Figure 2:** pyrometamorphic material at Ricetto (Italy). Image from [Stoppa *et al.* 2005](#)

261

Figure1_Stoppa2005_AM90_1919. MSA copyright.

262

263 2.1.3 Other processes fueling pyrometamorphism

264 It should be noted that pyrometamorphic rocks and glasses can be produced by high
265 temperature burning using any fuel, not just coal. Many examples have been described where
266 the fuel was biomass or organic material covered by sediment. This organic material can be
267 deposited naturally, and subsequently be covered to ignite and burn in the subsurface under dry
268 conditions, such as examples in Chile (Roperch *et al.* 2017) and Africa (Melson & Potts 2002;
269 Svensen *et al.* 2003). Alternatively, organic deposits can be a result of human activity, such as
270 archaeologically observed middens (Thy *et al.* 1995) or modern-day haystacks (Baker & Baker
271 1964), the burning of which produces glassy material.

272 Organic gases can be fuels for rock burning as well. One of the most studied areas of
273 pyrometamorphic rocks, the Dead Sea region in Israel, has been interpreted as having a
274 pyrometamorphic origin, from a mud volcano and associated methane and hydrocarbon gas
275 combustion (Sokol *et al.* 2010). Gas seeps and their associated ‘eternal flames’ from
276 spontaneous combustion have played important roles in human history, especially in religious
277 and mythological traditions. For example, the methane seeps known as Chimera in southwest
278 Turkey was the site of the first Olympic fire in the ancient Greek world, and the site of a temple
279 of Hephaestus (Vulcan) the god of fire and metallurgy (Etiope 2015; Hosgormez *et al.* 2008),
280 and it still burns today. All kerogens of various grades, from bitumen to oil, are potential sources
281 of fuel for creation of pyrometamorphic rocks and glass.

282 Pyrometamorphic melts are also known to be produced from oilfield fire. Characteristic
283 glasses, known as tengizites after the Tengiz oilfield in Kazakhstan, formed in this inferno
284 (Kokh *et al.* 2016). Tengizites, essentially a type of paralava/slag, contain 59-69 wt.% SiO₂,
285 7.3 - 9.7 wt.% Al₂O₃, 12.8 - 17.9 wt.% CaO, 2.0 - 3.7 wt.% MgO, 2.0 - 3.0 wt.% Na₂O, 1.3 -
286 1.9 wt.% K₂O, < 0.3 wt.% H₂O and < 0.4 wt.% total volatiles, as a result of melting sand and
287 clayey silt along with biogenic calcite (Kokh *et al.* 2016). The associated buchite at the site is
288 known as mesolite, consisting of partially melted and highly altered sediment, and compared to

289 tengizite has lower SiO₂, higher Al₂O₃, MgO, K₂O, TiO₂, and total volatiles, but lower
290 FeO/Fe₂O₃ ratio.

291

292 **One detailed example**

293 Compositions of pyrometamorphic glasses vary drastically depending on the local
294 chemical environment derived from the protolith, extent of melting, mixing of the melt, and
295 subsequent crystallization. For example, most Western US coal-bearing regions are interbedded
296 with predominantly shale and sandstone (Rogers 1918). The famous example Grimes Canyon
297 in California consists of protoliths of mudstone containing clays, detrital quartz and feldspar,
298 opal, apatite, biogenic carbonates (calcite and dolomite), and gypsum, while also being
299 bituminous, containing more than 10% carbon (Bentor *et al.* 1981). Some diatomites, shales,
300 and phosphorites also were locally metamorphized; phosphorite contains predominantly detrital
301 apatite, which melts at 1650 °C (Bentor 1984). Some constituents elevated in the
302 metamorphized rocks such as sulfate, and in other cases chloride, likely came from groundwater
303 introduction during the combustion process (Bentor *et al.* 1981; Rogers 1918).

304 Bentor (1984) distinguishes between combustion ‘glasses’ and ‘crystalline slags’ for
305 these materials, depending on the peak melting temperature achieved. ‘Glasses’ result from low
306 temperature melts ≤ 1000 °C, are enriched in Na, K, Al, and Si, and maintain high viscosity in
307 the small volume seams between unmelted rock, thus preventing crystallization. ‘Crystalline
308 slags’ melted at ≥ 1100 °C, dissolving even apatite crystals and other refractory minerals,
309 resulting in a lower viscosity, more basalt-like melt, which underwent extensive crystallization
310 on cooling (hence ‘slag’). These ‘slags’ are essentially synonymous with paralavas. There is
311 strong evidence of silicate/phosphate liquid immiscibility from the lower temperature silicate
312 melt and higher temperature phosphorite melt in these cases (Bentor 1984; Bentor *et al.* 1981;
313 Capitanio 2005; Peretyazhko *et al.* 2018).

314 Two samples of Grimes Canyon were investigated by X-ray diffraction. Not all phases
315 could be accurately identified by XRD, but diffraction indicated the majority was amorphous
316 glass (50-75%), with large fractions of feldspar (12-25%), apatite (6-7%), calcite (3-4%),
317 pyroxene (1-9%), and tridymite (1-4%). The lighter portions of the banded material contained
318 more carbonate and phosphate crystalline phases. These phases are broadly consistent with
319 those observed in [Bentor et al. \(1981\)](#). In Wyoming Powder River Basin paralavas, phenocrysts
320 of olivine, cordierite, and spinel (magnetite-hercynite-ulvospinel) are also common, along with
321 many unusual minerals and mineral associations ([Cosca et al. 1989](#)). In the Apennine region in
322 Italy, pyrometamorphic outcrops include wollastonite and melilite, likely from protoliths of
323 mixed marly sediments of carbonates and shales ([Melluso et al. 2004](#)).

324

325 In summary, pyrometamorphic glasses and rocks are highly variable due to the varying
326 temperatures, mineral protoliths, and local environmental conditions, resulting in equally
327 variable disequilibrium melting, volatilization, mixing, and crystallization ([Canil et al. 2018](#);
328 [Peretyazhko et al. 2018](#); [Sokol et al. 1998](#)).

329

330 **2.2 Pseudotachylite or frictionites**

331 Pseudotachylite is a generic name for friction melts, and usually, in the geological
332 terminology, pseudotachylites are classified as fault rocks ([Killick 2003](#)) even if the genesis
333 could be quite broad. In agreement with [Heide & Heide \(2011\)](#), we classify these glasses as
334 metamorphic ones, and in turn, among those glasses that originated from pre-existing rocks and
335 display marked modifications due to changes in the pressure-temperature conditions.

336 Friction melts (and especially those with a pumice texture) were firstly associated
337 exclusively to volcanic origin or to impact events. The term itself was introduced to describe
338 the abundant dark glassy veins in Vrederfort (South Africa), the largest impact structure on

339 Earth. Nowadays, pseudotachylites are associated to large impact structures, earthquake-
340 generated layers (veins), and very large rock avalanches (*e.g.* [Reimold & Gibson 2005](#); [Spray](#)
341 [2010](#)). Indeed, the exclusive volcanic or impact-associated origin hypotheses have been both
342 ruled out from the study of pumiceous rocks from Ötz Valley (Köfels landslide, see [De Blasio](#)
343 [& Medici 2017](#); [Erismann *et al.* 1977](#)), and since then, several studies focused on fault related
344 friction melts that form by co-seismic friction (fault pseudotachylite) or avalanches.

345 The typical generic definition provided for pseudotachylite is: “dense rock produced in
346 the compression and shear associated with intense fault movements, involving extreme
347 mylonitization or partial melting,” ([Bates & Jackson 1987](#)). Thus, the term “pseudotachylite”
348 should be used to describe rocks that present unambiguous evidence of high temperatures.
349 Because sometime this name is used regardless the origin of the glass, it is recommended to
350 point out the differences between impact-related and fault-related pseudotachylites, such as the
351 thickness of the material, or their formation history ([Kenkmann *et al.* 2000](#); [Reimold & Gibson](#)
352 [2005](#)). The mechanisms of formation of (impact)-pseudotachylites in large impact structure are
353 described in [Reimold & Gibson 2005](#). In the following we will briefly describe pseudotachylites
354 associated to very large landslides or formed along a fault plane during seismic deformation.

355 Fault pseudotachylite usually are veins of a few millimeters to a few centimeters in
356 width that are related to large magnitude seismic events and located mostly in the upper crust
357 ([Ferré *et al.* 2005](#)). Large seismic events can induce to a local increase of the temperature and
358 to the melting of the host rocks along the fault plane, resulting in the creation of amorphous
359 (cryptocrystalline) veins in response to frictional fusion. Depending on the maximum
360 temperature achieved during seismic slip, the mineral phases present in the host rocks will be
361 either melted or preserved as relict grains in the glassy matrix ([Spray 1992](#)). The chemical
362 composition of the fault pseudotachylites is often slightly more mafic than the host rock, but
363 the glass chemistry is dependent on bulk composition, mineral assemblage, and texture of the

364 host rocks (Di Toro *et al.* 2005; Maddock 1992). The study of the microlites and of the
365 compositions of the glass matrix can be used to determine the temperature regimes associated
366 to the seismic events. For example, artificial pseudotachylites have been produced by direct
367 high-speed friction experiments (*e.g.* Kenkmann *et al.* 2000; Lin & Shimamoto 1998; Spray
368 1987) and the estimated melt temperatures of natural and experimental pseudotachylites are in
369 the range of 750–1400 °C (Lin & Shimamoto 1998). Heide & Heide 2011 report melting
370 temperatures of 1700 °C because of the presence of lechatelierite inclusions in frictionite melts.
371 Nestola *et al.* (2010) report the occurrence of hexagonal $\text{CaAl}_2\text{Si}_2\text{O}_8$ (dmisteinbergite) in a fault
372 pseudotachylite from the Gole Larghe Fault (Adamello, Italy), a polymorph that is formed at
373 high temperatures (1200-1400 °C) by rapid cooling. The constrain of the temperature regimes
374 during seismic events is considered very important in order to better understand fault processes
375 and to infer earthquake source parameters.

376 Friction melts can be generated as well during very large landslides (Erismann *et al.*
377 1977; Legros *et al.* 2000; Weidinger *et al.* 2014). Well-studied landslides-derived
378 pseudotachylites are in Köfels (Austria) and Himalaya (Nepal). The latter is a giant rockslides
379 (dislocation of ~170 m) that caused the formation of a homogeneous glassy layer with thickness
380 between 1 - 3 cm (Masch & Preuss 1977). Masch & Preuss 1977 report a detailed study of both
381 events and they observed that glass matrix is chemically heterogeneous with schlieren, bubbles,
382 and relicts from the parent rock materials (partial to almost complete melting of host rocks of
383 granitic to granodioritic composition). Moreover, they report the occurrence of glasses with
384 pure quartz, plagioclase and alkali-feldspar compositions. Weidinger *et al.* (2014) report a
385 review of several basal deposits of giant rockslides and defined that very short (<10 s) giant
386 moving rockslides could produce friction-induce partial melting with basal temperatures > 1500
387 °C (in anhydrous conditions).

388

389 **2. Glasses from highly energetic events**

390
391 We will describe amorphous materials deriving from highly energetic events, either
392 natural or artificial. First of all, glasses produced by natural events, such as tektites and impact
393 spherules, will be taken into account. Then, we are going to describe some enigmatic natural
394 glasses that are almost certainly of impact origin (accepted or assumed) but that are not found
395 in stratigraphic contexts. Notable examples of such glasses are Libyan Desert Glass, Darwin
396 Glass, and Dakhleh Glass. Finally, we are going to describe other glasses, resulting from a rapid
397 “shock” event, the fulgurites. At the end, we will consider also a glass, not strictly natural, but
398 that can be considered as an analogues of impact glasses: the atomic glass - trinitite.

399

400 **2.1 Impactites**

401 Impact processes are able to greatly influence the evolution of planets. Just looking at the
402 Moon, we can recognize with bare eye that the surface is heavily affected by impact cratering.
403 Besides our satellite, planets such as Mercury, Mars and many satellites in the Solar System
404 have circular depressions that testify the occurrence of numerous impacts, with only a few
405 exceptions, including Earth, where the topography has been remodeled by active geological
406 processes. The first impact crater directly witnessed on Earth (the so-called Carancas event) is
407 very recent. It occurred near the southern shore of Lake Titicaca in Peru on September 2007,
408 where the impact of a stony meteorite produced a 13.5 m crater ([Tancredi et al. 2009](#)).

409 The recognition of impact cratering as a fundamental process in the evolution of a planet
410 has occurred as well rather recently, with the discovery of small spherules worldwide
411 distributed in the Cretaceous- Paleogene K-Pg layer (formally known as KT, Cretaceous-
412 Tertiary), first detected in Gubbio, Italy by [Alvarez and coauthors \(1980\)](#). These spherules and
413 the associated layer contain chemical and physical evidences for a major impact that caused the
414 mass extinction event at the K-Pg boundary. In 1991, a 170-180 km diameter crater was

415 identified on the Yucatan peninsula as the impact site (Chicxulub crater) of a 10 to 15 km
416 asteroid that induced the cataclysmic event responsible for the formation of the K-Pg worldwide
417 ejecta horizons and caused the end-Cretaceous mass extinction, around 65 Ma ago.

418 As might be expected, impacts have caused catastrophic effects on climate and landscape,
419 however, they do not produce just destruction and extinction. Indeed, there are also beneficial
420 effects associated to such events, such as the economic significance of impact deposits (*i.e.*
421 Sudbury, Canada; Vredefort, South Africa), or the associated hydrothermal systems created
422 (Osinski & Pierazzo 2012; Pierazzo & Melosh 2012).

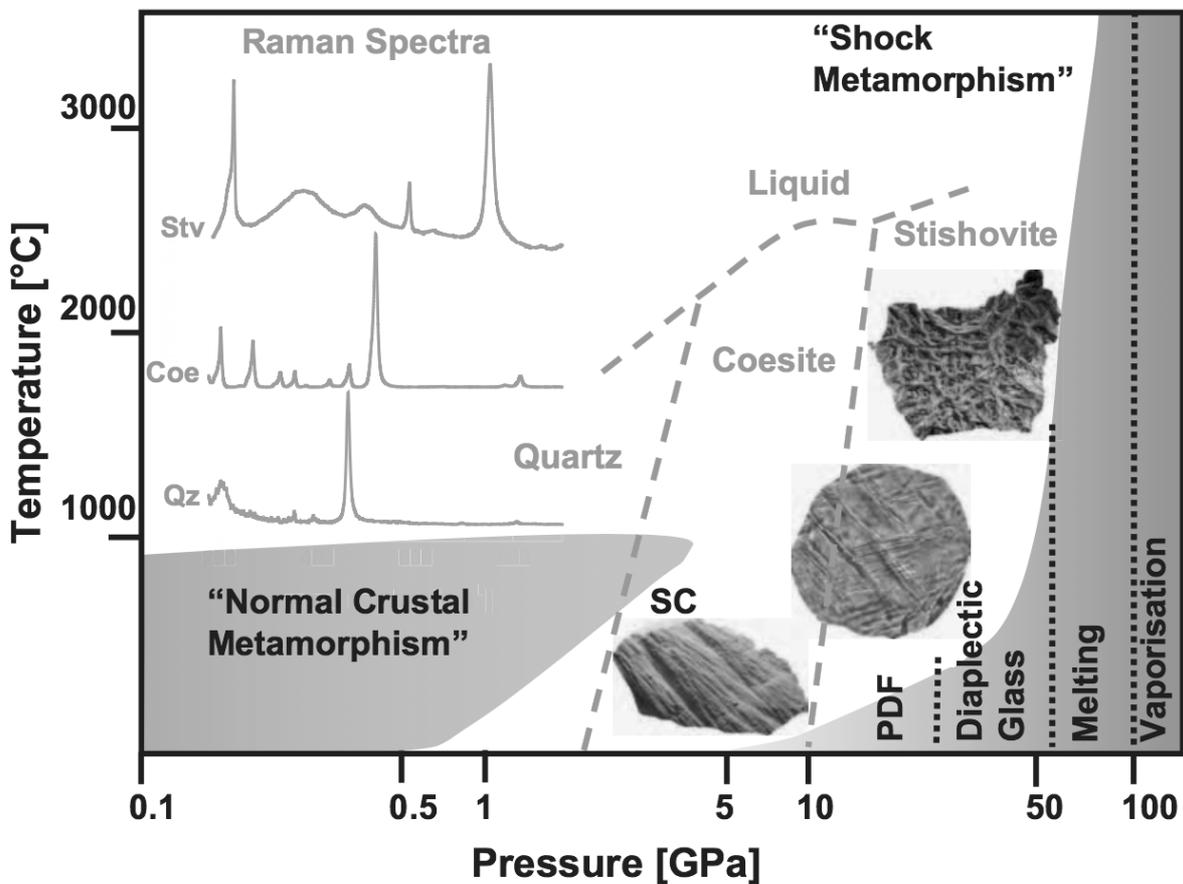
423

424 The impact of a large object (>50 m for a stony object), moving at a great speed (> 11 km
425 s^{-1} ; French 1998), with the surface will create an impact crater after a very short sequence of
426 complex events, which can be summarized into three main stages: contact and compression,
427 excavation and modifications (Melosh 2011). The generic term impactite refers to the large
428 variety of materials (rocks, melts) formed by hypervelocity impact(s) of a large extraterrestrial
429 body. A series of distinct materials is generated by the forces of a hypervelocity impact, starting
430 with a shock wave (compression stage) and followed by decompression from peak shock
431 pressures, with associated heat generation and material transport (excavation stage) Reimold &
432 Jourdan 2012. The post-shock heat causes the solid-state deformation and/or melting of the
433 target crustal rock and even its vaporization, and the original material is entirely altered by the
434 extreme heat and pressure regimes. Simulation of impact processes have established that the
435 timescale of crater formation is extremely short, compared to many other geological processes,
436 and are in the order of seconds to minutes for craters ranging from 1 to 100 km, thus impactites
437 will present very distinctive characteristics (French 1998; Stöffler *et al.* 2018; Stöffler & Grieve
438 2007). Impactite is the commonly used (and misused) word for all shocked, melted or vaporized
439 materials as a consequence of a hypervelocity impact, and because of the confusion, a study

440 group proposed a classification and a nomenclature of these materials, based on geological
441 setting, texture, composition, and degree of shock metamorphism (Stöffler *et al.* 2018; Stöffler
442 & Grieve 2007). The first grouping is based on the location of the materials, thus “proximal
443 impactites” is used for materials found in the proximity of the impact crater, whereas the term
444 “distal impactites” refers to materials that do not occur directly in or around a source crater (*i.e.*
445 distal ejecta). Further sub classifications have been applied to proximal impactites, and
446 according to Stöffler & Grieve 2007, proximal can be further divided into three subgroups:
447 shocked rocks, impact melt-rocks and impact breccias; depending on the peak pressure and the
448 post-shock temperatures, both minerals and original rocks texture will present shock effects
449 that allow to identify progressive stages of shock metamorphisms. For instance, by increasing
450 shock pressure there are i) Planar Fractures (PFs) and Planar Deformation Features (PDFs) in
451 minerals such as quartz, zircon and feldspars; ii) high-pressure quartz polymorphs (coesite,
452 stishovite); iii) diaplectic mineral glasses (produced without fusion); iv) fused mineral glasses
453 (produced with fusion), and v) melts (see **Figure 3** from Reimold & Jourdan 2012). For
454 example, quartz grains exposed to shock compression develop planar microstructures
455 depending on the pressure range: PFs for pressure range of 5 - 8 GPa, and PDFs over the
456 pressure range of 5-10 GPa to ~ 35 GPa. Shock-induced deformation in zircon grains have been
457 reported as well, indicating shock pressures higher than 20 GPa (*L. Ferrière and G. R. Osinski –*
458 *impact cratering book, chapter 8*). Impact diamonds have been reported as well in many impact
459 sites. The transition from carbon (graphite) to diamond and lonsdaleite is around 13-15 GPa,
460 with temperature in the range of 1300–2000 K (French 1998; Pratesi 2009).

461 **Figure 3** clearly shows that the pressure conditions for normal crustal metamorphism
462 (regional, contact) differs completely from shock metamorphism, resulting in the production of
463 characteristic features and materials. In normal crustal metamorphism the range of temperature
464 and pressure is around ≤ 1000 °C and 1 - 3 GPa, respectively, and rocks and mineral

465 transformations occur in very long times ($>10^5$ years), thus approaching equilibrium (French
 466 1998). On the contrary, shock metamorphism is an instantaneous process, with peak pressures
 467 that reach > 100 GPa, and temperatures much higher than 3000 °C. Because of the rapid process,
 468 quenched amorphous and crystalline metastable phases are characteristic. For a detailed
 469 description of all shocked materials, readers are referred to the comprehensive work of Stöffler
 470 & Langenhorst 1994 and to Osinski *et al.* 2012).



471
 472 **Figure 3 - Pressure (P)-temperature (T) diagram, for normal and shock metamorphisms.**
 473 **Figure from Reimold & Jourdan 2012; COPYRIGHT MSA**
 474

475 In the following, we will mainly discuss the most homogeneous glasses created by
 476 hypervelocity impact events, *i.e.* the distal ejecta tektites, but we will also provide a brief
 477 overview of some other distal ejecta and proximal glasses. To distinguish proximal impact
 478 glasses, volcanic glasses and distal ejecta tektites, Koeberl 2013 provides some characteristics
 479 that are distinctive of the distal ejecta, and enable their recognition. Tektites are:

- 480 i) amorphous and fairly homogeneous (no crystallites);
481 ii) contain lechatelierite (amorphous SiO₂);
482 iii) occur within definite areas, called strewn-fields (SF) and are associated to a single
483 source impact crater;
484 iv) do not occur directly in or around a source crater (they are distal ejecta).

485 Moreover, tektites are strongly depleted in water (0.002 to 0.02 wt.%, at least an order of
486 magnitude lower than the H₂O content of volcanic glasses) and are highly reduced (almost all
487 iron occurs as Fe²⁺) (Beran & Koeberl 1997; Fudali *et al.* 1987; Giuli *et al.* 2010a, 2013a,
488 2014a; Giuli 2017; Koeberl 1994, 2013; Melosh & Artemieva 2004; Rossano *et al.* 1999).

489

490 3.1.1 Distal Ejecta

491 As a consequence of a hypervelocity impact, the uppermost (200 m) surficial target rock
492 is melted and ejected. A small portion of this molten ejected material will fly far away from the
493 source crater and are called distal ejecta (according to Montanari & Koeberl 2000, ~ 90 % of
494 the ejecta are proximal *i.e.* deposited within five crater radii from the impact site). The flying
495 melts are quickly quenched to form fairly homogeneous amorphous materials with typical
496 shapes (*e.g.* splash forms) like spheres, teardrops, disc-shaped form, and are known as tektites.
497 A partial crystallization of primary microlites/crystallites (*e.g.* clinopyroxene or Ni-rich spinel)
498 upon cooling could occur and the material formed is called microkrystite (Glass & Simonson
499 2013 and references therein).

500 Distal ejecta deposits (or air-fall beds) are deposited far away from the source crater and
501 can be divided in two types: i) tektites and microtektites, and ii) spherule beds (Osinski *et al.*
502 2012). A more enigmatic kind of distal ejecta found on Earth consists of glassy materials, not
503 volcanic in origin, but not having spherical shapes and not classified as tektites. Example of
504 these enigmatic glasses are Libyan Desert Glass, and Darwin glass (Glass & Simonson 2013).

505 The reasons behind the formation of spherules and tektites remains poorly understood, and no
506 solid model or evidence has been found to explain why just a few impacts form large amounts
507 of high-speed ejected glasses and why only a few provides tektites ([Howard 2011](#)). Thus, tektite
508 formation must require specific impact conditions (see below).

509 On Earth, impact craters are not easily found (190 confirmed impact structure as of July
510 2019; [Earth Impact Database 2019](#)) because their morphology has been altered by hydrothermal
511 and chemical alteration, and by tectonic processes. Thus, most of the impact craters occurred
512 on Earth have been destroyed or covered. For instance, the Chicxulub impact structure is
513 located beneath ~ 1 km of sediment below and half offshore the Yucatan Peninsula, and until
514 1991, year of the discovery, the only proof of a connection between end-Cretaceous mass
515 extinction (K–Pg boundary layer) and a possible cataclysmic impact, was the worldwide
516 occurrence of distal ejecta horizons. Hence, sometimes, distal ejecta are the only remaining
517 witnesses of large impact events, and they provide important information regarding planetary
518 processes when found in the stratigraphic record. The importance of air-fall beds has been
519 compared to the study of volcanoclastic layers ([Glass 2016](#)).

520 When dealing with distal ejecta, or in general, with glass spherules, the most important
521 point is to gather enough evidences to rule out the volcanic origin. These evidences are usually
522 called ‘impact markers’ and includes chemical, isotopic, and mineralogical marks that indicate
523 the involvement of a cosmic body. Example of markers that indicate an extraterrestrial
524 contribution are the elevated content of siderophile elements (Ir, platinum group elements,
525 PGE), the occurrence of shocked minerals a/o Ni-rich spinels a/o impact diamonds, and atypical
526 isotope ratios. The complete list of the well-established or new ‘impact markers’ is beyond the
527 scope of this chapter, and the readers are referred to the exhaustive contribution of ([Goderis et](#)
528 [al. 2012](#)).

529

530 **Tektites and microtektites**

531 Tektites are small, typically dark, glassy objects that have been transported through the
532 atmosphere hundreds to thousands of kilometers from the impact site, and are found only in
533 certain areas of the Earth's surface, called tektite strewn fields (SF). Tektites are generally
534 chemically homogeneous, Si-rich glasses of various sizes (usually > 1 cm), with typical
535 aerodynamic shapes and very characteristic surface features (Figure 4). Microtektites are
536 microscopic tektites, with diameter < 0.1 cm, largely found in deep-sea sediments.

537 Tektites can be further subdivided in three types, depending on their forms: Muong Nong-
538 Type (or layered), ablated/flanged tektites (or aerodynamically shaped tektites), and the most
539 common ones, the splash forms (or normal tektites). The morphology of the most common
540 tektites varies between spherical, dumbbell-shaped, and teardrops (Figure 4) as a result of rapid
541 rotation of the molten flying material.



542 **Figure 4 - Australasian tektites with various shapes; dumbbell, teardrop and fragments. Dumbbells**
543 **are formed by rapid rotation of melt droplets ejected into the atmosphere. When the rotation is high**
544 **enough, the dumbbell is broken to form teardrops.**
545
546

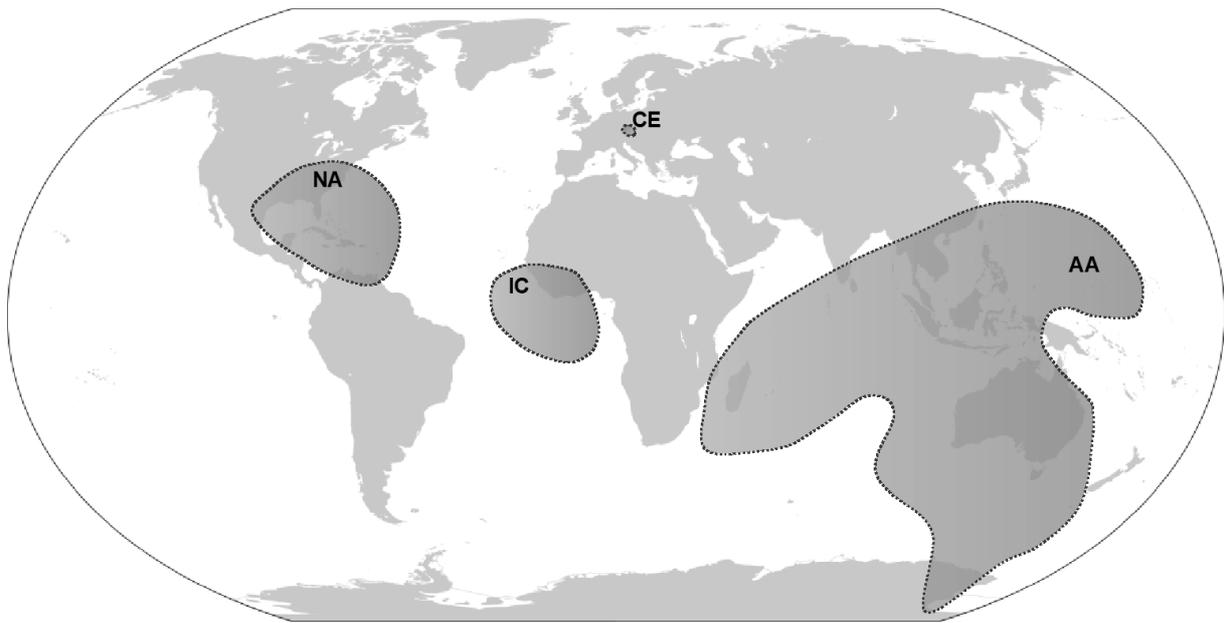
547 Muong Nong-Type tektites (MN - named after a region in Laos) are unusually large
548 tektites (up to several tens of centimeters in size) and with layered structures. [Koeberl 1992a](#)

549 observed that Muong Nong-Type tektites are enriched in volatile elements (the halogens, Cu,
550 Zn, Ga, As, Se, Pb), and have higher water contents compared to splash-forms and ablated
551 tektites. Moreover, MN tektites present chemical heterogeneity (darker and lighter layers), and
552 may contain relict mineral grains (*e.g.* corundum, quartz, chromite and cristobalite) and very
553 large bubbles (Glass & Simonson 2013). Despite the differences with other tektites, still MN-
554 Type tektites have an average chemical composition (major element chemistry) and age
555 matching those of common tektites. Moreover, MN-Type tektites strongly differ from volcanic
556 glasses because of the presence of shocked mineral inclusions, for differences both in major
557 and trace element contents (*e.g.* REE patterns), lower water content, and highly reduced iron
558 (Koeberl 1992a). The presence of relict mineral grains, and the relatively higher water amount
559 in Muong Nong-Type tektites suggest that these glasses have experienced the lowest
560 temperatures of all tektites (Koeberl 1992a). For instance, the presence of volatiles (halogens,
561 Zn) and the vesicularity of lechatelierite particles seem to indicate lower formation
562 temperatures (Montanari & Koeber 2000). It is assumed that Muong Nong-type tektites have been
563 deposited closer to the source crater and based on ^{10}Be data they should derive from a greater
564 depth in the target deposits than were most of the other tektites (Ma *et al.* 2004). The layered
565 structure of these tektites, with their chemical heterogeneities, and the presence of relict mineral
566 grains, may serve to better constrain tektite conditions of formation, because they supposedly
567 represent the major link between target rocks and tektites (Montanari & Koeber 2000).

568 To date, of the four known tektite strewn-fields, all but one have been linked to source
569 craters, based on geographic location, geochemical evidences, and composition (Koeberl 2013
570 and references therein). The oldest strewn field known is the North American (NA) one of 35.3
571 Ma age (± 0.1) associated with the 85 Km diameter Chesapeake Bay (USA) impact structure
572 and includes Bediasites, Georgianites, Barbados and Cuba tektites. The central European (CE)
573 or moldavite strewn field of 14.8 Ma age (± 0.2 ; Schmieder *et al.* 2018) is associated with the

574 Nördlinger Ries impact structure of about 24 km in diameter (Nördlinger Ries, Bavaria, D).
575 There is another impact crater, the Steinheim crater, ~ 3.8 km in diameter, located about 42
576 kilometers west-southwest from the center of Nördlinger Ries. These two craters are believed
577 to have formed nearly simultaneously by the impact of a binary asteroid (Stöffler *et al.* 2002).
578 Moldavites are found mainly within a 200–450 km ranges from the Ries crater, in Czech
579 Republic (Bohemia, Moravia), Austria and Germany. Recently, a sub-strewn field in Poland
580 has been reported as well (Brachaniec *et al.* 2014.), circa 485 km from the Ries crater.
581 Moldavites have a unique green color and have strong surface sculpture deriving from erosion.
582 Stöffler *et al.* 2002, and more recently Artemieva *et al.* 2013, modeled the formation of the CE
583 strewn field, suggesting a 1.1-1.5 km stony meteorite impactor hitting the target surface with
584 an angle of 30°, at 15-18 km/s. The Ivory Coast (IC) tektite strewn field is associated with the
585 1.07 Ma old Bosumtwi crater (10.5-11 km diameter, Ghana, Africa) (Koeberl *et al.* 2007). The
586 Bosumtwi crater is the youngest well-preserved impact structure known, and it is almost
587 completely filled by a lake (Lake Bosumtwi). The youngest strewn-field, of 803±3 ka (Lee &
588 Wei 2000), is the Australasian one, for which no source crater has been identified so far.
589 Tektites of the Australasian strewn field (AA) include australites, thailandites, indochinites,
590 philippinites and javanites. The AA strewn field is very extensive and finding of microtektites
591 on land and oceans (microtektite-bearing deep sea cores) have improved the determination of
592 the AA geographic distribution, which is inhomogeneous (Figure 5) and spread from the
593 southeastern region of Asia down to Australia and Tasmania. Recently, smaller tektites
594 (microtektites) that were found in four different sites in northern Victoria Land Transantarctic
595 Mountains (TAM, Antarctica) have been related to AA and there are clear evidences Folco *et*
596 *al.* 2008, 2009, 2010, 2011; Giuli *et al.* 2014a) that these samples represent a major
597 southeastward extension of the Australasian strewn field. Thus, the AA strewn field covers
598 more than 10% of the Earth's surface, and even if no source crater has been identified yet,

599 several authors suggest its location somewhere in the Indochina region (Folco *et al.* 2016).
600 Besides these four well-known tektite strewn fields, recently, other two have been proposed,
601 one from central American (Belize glasses), and the second from Uruguay (uruguaites)
602 (Ferrière *et al.* 2017; Giuli *et al.* 2014b; Povenmire *et al.* 2011; Povenmire & Cornec 2015;
603 Senftle *et al.* 2000).
604



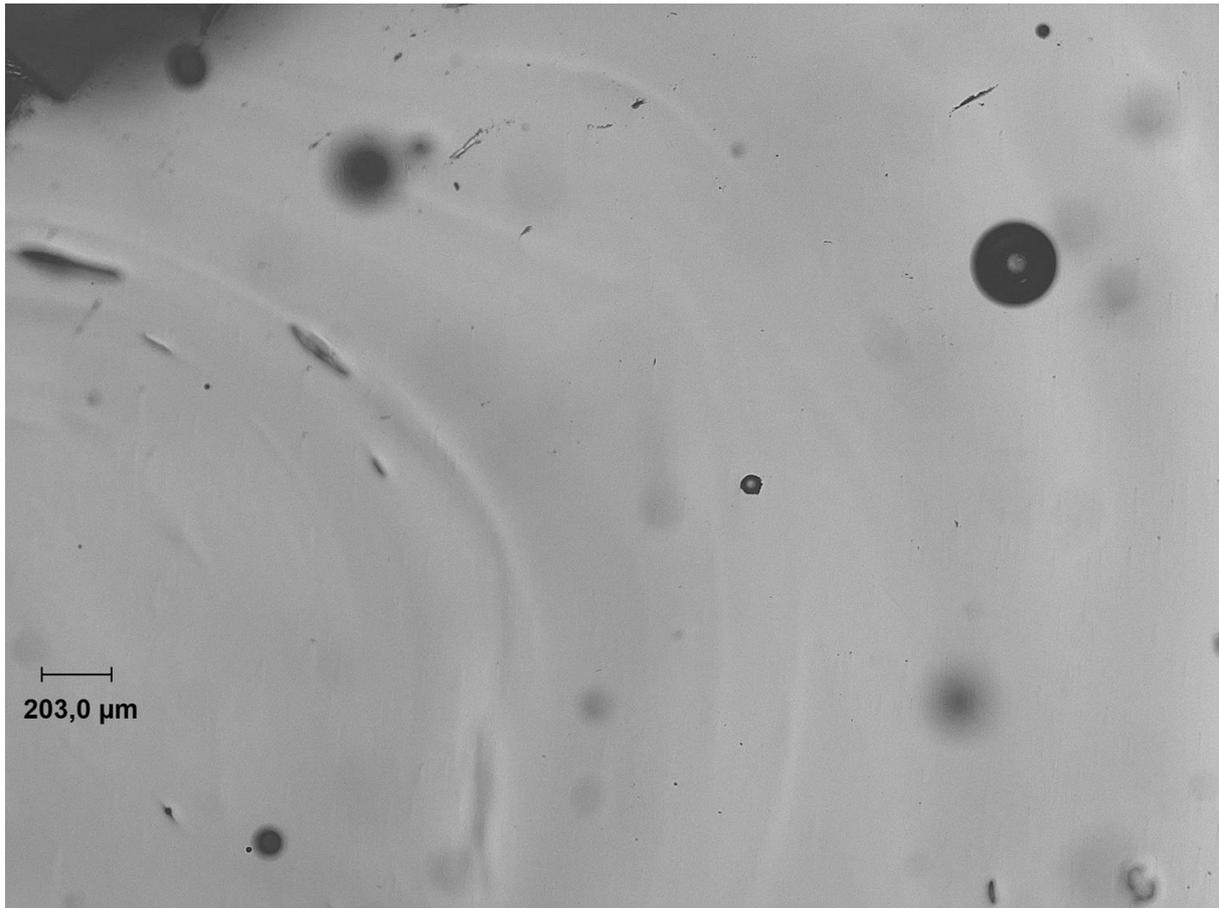
605
606 **Figure 5:** Approximate location and extension of the four strewn fields: NA (North American), IC (Ivory
607 Coast), CE (Central Europe) and AA (Australasian). The known source craters are Chesapeake Bay (NA),
608 Ries (CE) and Bosumtwi crater (IC). Redrawn after Folco *et al.* 2011, 2016; Glass 2016.
609

610 The chemical composition of tektites is fairly homogeneous within the same strewn-field,
611 and also among the four strewn-fields the chemistry of all distal ejecta shows that these glasses
612 are aluminosilicates with high SiO₂ contents and alkalis depleted (see Table 1).

613 Tektites are usually black/dark green, except moldavites that have a characteristic lighter
614 green color, and do not contain microlites or relict minerals, but they contain vesicles, mostly
615 spherical, or elongated as evidence of glass flow. Figure 6 shows the light transmitted optical
616 image of an australasian tektite. Flow lines and small vesicles are clearly visible. Bubbles are
617 very frequent in tektites, and many studies have been done in order to determine the

618 composition, the content and pressure of the trapped gases. These data are very important
619 because they provide information on the nature of the atmosphere during formation
620 (quenching). Oxygen, C-based (CO, CO₂, CH₄) and noble gases have been identified in
621 different proportions, and sometimes even contradictory results have been presented, mainly
622 because of the extraction methods used. Early studies reported the presence of CO₂, CO, H₂,
623 SO₂, CH₄, N₂, and O₂ with total gas contents ranging between 0.23 and 0.82 cm³/g ([Ottemann
624 1966 and references therein](#)). [Gentner & Zähringer \(1959\)](#) and [Zähringer & Gentner \(1963\)](#)
625 studied noble gases in several tektites and found heavy noble gases (Ar: Kr: Xe) ratios very
626 similar to that of the Earth's atmosphere, thus confirming the terrestrial origin of these
627 specimens. Moreover, the established contents of argon (Ar⁴⁰) was fundamental to determine
628 the age of tektites across the AA strewn fields and to confirm that the different specimens were
629 of the same age, and in turn, of the same origin. A recent review by Zak et al (2012) lists all
630 studies concerning C-based gases in tektites (excluding noble-gases). More recent studies on
631 gas ratios, and noble gas isotopic ratios confirmed that gases have terrestrial atmosphere origin,
632 with a typical enrichment of lighter noble gases (He, Ne) compared to the heavy ones (Ar, Kr,
633 Xe) (*e.g.* [Jessberger & Gentner 1972](#); [Matsubara & Matsuda 1991](#); [Matsuda et al. 1996](#); [Müller
634 & Gentner 1968](#); [O'Keefe et al. 1962](#)). Moreover, the estimated bubbles internal pressures are
635 very low (\ll 1atm) thus suggesting that vesicles formation must result from internal gas
636 pressure during tektite cooling, that happened in an atmosphere under low pressures (at high
637 altitude). For instance, [Matsuda et al. \(Matsuda et al. 1996\)](#) reported a bubble pressure of $\sim 10^{-4}$
638 atm, and a solidification of the molten ejecta that should have occurred high in the stratosphere
639 at altitudes above 20-40 km, in agreement with earlier observations ([Rost 1964](#)).

640



641

642 **Figure 6** – Optical microscope image (transmitted light) of an Australasian tektite slice. Several bubbles
643 of different dimensions, and flow lines are visible.
644

645 Three of the four tektite strewn-fields so far known also present microtektites (diameter
646 usually < 0.1 cm): the North American, the Ivory Coast, and the Australasian SF. These
647 spherules have been found in deep-sea deposits (see, *e.g.* [Glass 1967, 1972, 1978, 1990](#)) and
648 are very important for defining the extension of the strewn fields (*e.g.*, [Glass & Zwart 1979](#)),
649 for constraining the stratigraphic age of tektites, and to provide an indication regarding the
650 location of possible source craters (*e.g.*, [Glass & Pizzuto 1994](#)). Ivory Coast microtektites are
651 found as far as > 2000 km away from their source, whereas Australasian microtektites have
652 been found also at extraordinary distances (>10000 km) from the hypothetical source crater.
653 Microtektites, like tektites, show various morphologies, with oblate/prolate spheres, dumbbell-
654 and teardrop-shaped, and may contain lechatelierite and vesicles. However, microtektites
655 display a larger range of colors than tektites, with the majority being transparent, colorless, or

656 greenish/yellowish. Trace elements and isotopic abundances confirm that microtektites are
657 genetically related to tektites in the associated strewn field (Frey 1977), but microtektites
658 usually show a wider compositional range than tektites, even if, generally, they are similar to
659 the tektites from the same strewn-field (Table 1). For example, it is possible to recognize IC
660 and AA microtektites based on their alkali earth ratios (MgO/CaO).

661 There is a particular sub-group of AA microtektites that presents really high Mg
662 contents (HMg; up to 24 wt.% MgO) and low SiO₂ contents than “normal” AA-microtektites,
663 and that have bottle-green color and are highly eroded (Glass & Simonson 2013). A detailed
664 study of major and trace elements of >100 microtektites from the different strewn fields have
665 been done by Glass *et al.* (2004) that besides the HMg microtektites, found also some glasses
666 with high Ni contents (HNi; up to ~470 ppm). HMg microtektites have been found also in the
667 IC strewn-field. The study of the trace elements in microtektites also confirmed that these small
668 distal ejecta derive from upper continental crust.

669 Everything seemed to point to microtektites being just smaller tektites. However, lately
670 some differences have been found, namely on the Fe oxidation state of North American
671 microtektites (Giuli *et al.* 2013a). The authors have shown that some North American
672 microtektites present higher Fe³⁺/Fe²⁺ ratio (up to 0.61), compared to the respective tektites,
673 implying that, probably, different formation mechanisms are involved for the formations of
674 such small objects. Interestingly, for these microtektites there is a positive correlation with the
675 distance from the known source crater, and more oxidized conditions are reported for longer
676 distances. This seems to be in contrast with the data available on other microtektites recovered
677 at much further distances (*i.e.* AA microtektites). Because of the relative limited information
678 available on tektite/microtektite formation mechanisms (see below) it is difficult to provide an
679 interpretation of those data. Do microtektites register another “path” of the ejecta, or perhaps a
680 different timeframe? Do they have different cooling rates? Further experimental studies, along

681 with modelling and further ejecta discoveries will help answering those questions, and probably
682 the broader question of “how do distal ejecta form?”.

683

684

685 **Table 1: Average major oxide compositions of tektite and microtektites (wt.%). Data from § Glass**
686 **2016; ‡ Giuli *et al.* 2013a; # Žák *et al.* 2012; \$ Mizera *et al.* 2016; * Giuli *et al.* 2014a (TAM:**
687 **microtektites from Victoria Land Transantarctic Mountains); ⊕ Glass *et al.* 2004. For the original**
688 **data source and error, see the cited references.**

689

	NA §	NA micro ‡	CE #	IC §	IC micro ‡	AA \$	MN-type \$	TAM micro *	AA-HMg micro ⊕
SiO₂	81.80	73.50	80.30	67.58	67.37	73.70	78.00	72.27	54.73
TiO₂	0.51	0.68	0.32	0.56	0.54	0.78	0.62	0.88	0.92
Al₂O₃	11.20	14.08	10.10	16.74	16.68	12.30	10.60	15.07	17.41
FeO	2.64	3.99	1.69	6.16	6.46	4.35	3.81	3.77	6.02
MnO			0.10	0.06		0.10	0.08		
CaO	0.45	1.59	1.35	1.38	1.57	2.41	1.51	3.49	4.82
MgO	0.61	1.81	1.69	3.46	3.72	2.21	1.74	2.87	15.36
Na₂O	0.94	1.87	0.55	1.90	1.89	1.37	1.33	0.25	0.64
K₂O	2.44	2.24	3.48	1.95	1.77	2.78	2.18	0.93	0.11

690

691

692 Other distal ejecta

693 Besides tektites and microtektites, it is worth mentioning the occurrence of many other
694 spherule-associated layers across the world. For example, a clinopyroxene-bearing spherules
695 layer (cpx-spherules) has been traced in many locations on Earth. The cpx-spherules associated
696 layer is enriched in Ir and is ~10-20 ka older than the North American microtektite one (Glass
697 *et al.* 2004 and references therein). These spherules have been associated to the Popigai
698 complex crater (Siberia, Russia), an extremely large structure (~100 km diameter) of 35.7±0.2
699 Ma (Melosh 2011).

700 As mention before, the most notorious and studied distal ejecta deposits are the K-Pg
701 (former KT) spherule ones, associated to another gigantic impact, and to the 170-180 km
702 Chicxulub crater (Yucatan, Mexico). The impact event responsible for this crater had enough
703 energy to distribute ejecta worldwide, and cause the end-Cretaceous mass extinction, around

704 66 Ma ago. The small K-Pg spherules (100-500 μm), that resemble microtektites, were first
705 detected in the Cretaceous-Paleogene K-Pg layer in Gubbio (I) (Alvarez *et al.* 1980). K-Pg
706 distal impact ejecta horizons are associated with Ir enrichments, siderophile element anomalies,
707 and shocked minerals and high-pressure polymorphs (quartz grains, coesite and stishovite). K-
708 Pg spherule layers have a global geographical extension with more than 350 sites identified,
709 but because of the poor preservation of the claystone at the K-Pg boundary, in the early 80s
710 there were some debates on the origin of these spherules, with some authors supporting an
711 impact hypothesis (*e.g.* Smit & Klaver 1981), and others attributing an authigenic origin for the
712 spherules (*e.g.* Izett 1987). A few years later, many authors, by studying many K-Pg spherules,
713 and in particular the Si-rich spherules preserved at the K-Pg layer at Beloc (Haiti), provided
714 clear evidences of an impact origin, based on geochemical data, enrichment in platinum group
715 elements, the presence of lechatelierite and shocked quartz grains (Bohor 1990; Koeberl 1992b;
716 Koeberl & Sigurdsson 1992; Sigurdsson *et al.* 1991). Despite the alteration of some deposits,
717 according to Morgan *et al.* (2006) there is a correlation between shock markers and
718 paleodistances from the impact site, and in particular the number and size of the spherules and
719 shocked minerals are inversely proportional to their distance from Chicxulub. These evidences
720 have been used to confirm the occurrence of a single highly energetic impact event related to
721 the formation of the K-Pg spherule horizons, and to provide insights on the obliquity of the
722 projectile.

723 Among the proposed impact-related Cenozoic distal ejecta layers there is one horizon
724 related to a hypothesized impact event that according to Firestone *et al.* (2007), may have
725 occurred at the beginning of the Younger Dryas (YD ~12.8 ka). In this case, impact event is
726 not strictly related to a collision with the Earth surface, but (allegedly) to an airburst, that is a
727 shock wave caused by the explosion of a cosmic object in the atmosphere (such as the well-
728 known 1908 Tunguska airburst, Siberia). The Younger Dryas boundary is a < 5 cm-thick

729 sediment layer dated ~ 12.8 ka in several sites in North America, and in Belgium and Syria
730 (Bunch *et al.* 2012; Firestone *et al.* 2007; Wittke *et al.* 2013) that contains impact-related
731 markers, such as Ir enriched grains, magnetic micro-spherules (10-250 μm), vesicular carbon
732 spherules, glass-like carbon, shock-fused vesicular lechatelierite, impact nano-diamonds, and
733 fullerenes containing extraterrestrial concentrations of ^3He (helium-3 is extremely rare in
734 terrestrial crustal rocks; Koeberl 2013). Still, the YD impact remains just a hypothesis because
735 most of the reported “impact-related” markers could not be confirmed in independent studies
736 or are not considered unambiguously impact proxies. For instance, fullerene with anomalous
737 ^3He contents has not been found in all sites, and no shocked minerals have been reported.
738 Moreover, Paquay *et al.* (2009) measured both PGE concentrations and Os isotope ratio
739 ($^{187}\text{Os}/^{188}\text{Os}$) in the bulk sediments without finding evidences of extraterrestrial components.
740 In a review, Pinter *et al.* (2011) report that none of the evidences described by Firestone were
741 incontrovertible, and more recently, van Hoesel *et al.* (Van Hoesel *et al.* 2014), in another
742 review on the YD impact markers, concluded that no unambiguous markers have been found
743 so far, and that the data available still cannot support the claim that there was a Younger Dryas
744 impact event.

745

746 **Enigmatic Impact glasses**

747 Glass & Simonson (2013) in their contribution on distal impact ejecta describe some
748 glasses, almost certainly of impact origin, but not found in stratigraphic contexts. Most of the
749 time, no source craters could be associated to these materials, hence the melting has been often
750 attributed to radiation from airburst. These natural glasses have been found in several locations
751 on Earth and cannot be classified as tektites or spherules. Examples of such glasses are: glass
752 fragments from South Ural (Russia) called urengoites (Deutsch *et al.* 1997) glass fragments

753 from Egypt (Dakhleh glass; [Osinski et al. 2007](#)), and the famous Libyan Desert Glass, and
754 Darwin Glass (with a putative associated crater; [Meisel et al. 1990](#)).

755

756 In an area of ~ 6500 km² in southwest Egypt, close to the border with Libya, natural Si-
757 rich glass fragments, known as Libyan Desert Glass (LDG) are found ([Rocchia et al. 1996](#)).
758 Fission track dating provides an average age of ~ 29 Ma (28.5 ± 2.3 Ma to 29.4 ± 0.5 Ma
759 ([Storzer & Wagner 1977](#)), and 28.5 ± 0.8 Ma ([Bigazzi & Michele 1996](#)). LDG occurs as
760 centimeter- to decimeter-sized, irregularly shaped, and strongly wind-eroded glass pieces
761 ([Figure 7](#)), and one of the most famous specimens is the one used as the yellowish scarab of
762 Tutankhamen's pectoral. Libyan Desert Glasses are very silica-rich (~ 96.5–99 wt.% SiO₂; see
763 [Figure 1; Table 2](#)) and shows a limited variation in major and trace element abundances (the
764 remaining few wt.% are oxides of aluminum, iron, calcium, and magnesium). They are
765 homogeneous with a characteristic pale yellow/yellow-green color and characterized by the
766 presence of schlieren ([Glass & Simonson 2013](#)). Some minor elongated vesicles and spherulitic
767 inclusions of cristobalite have been reported as well, and in some cases baddeleyite (as high-
768 temperature breakdown product of zircon), rutile, staurolite, cordierite, kyanite, reidite and
769 mullite has been detected ([Cavosie & Koeberl 2019; Gomez-Nubla et al. 2017; Greshake et al.](#)
770 [2010, 2018; Rocchia et al. 1996; Storzer & Koeberl 1991](#)).

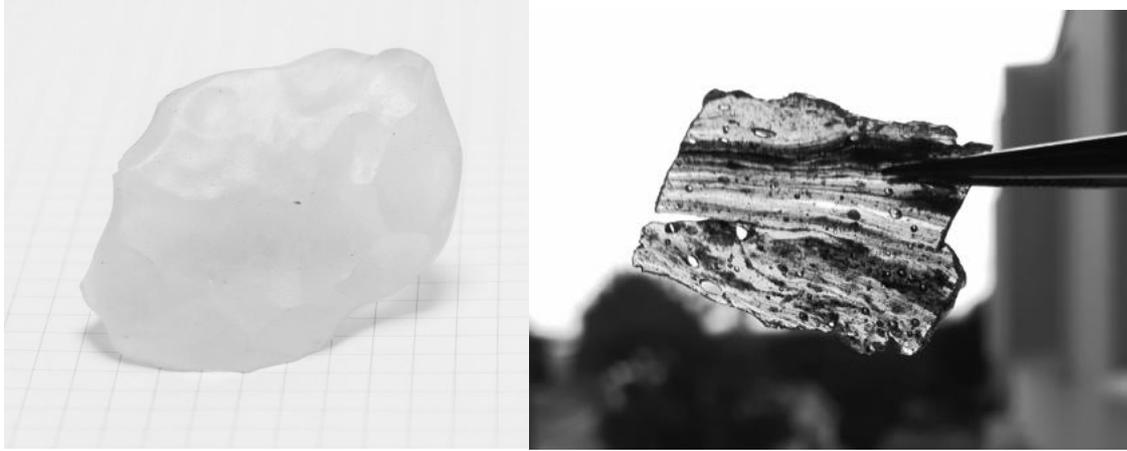
771 Since its discovery, early in the 20th century ([Clayton 1934](#)) the origin of the Libyan
772 Desert Glass still represents an unanswered enigma to all scientists and researchers. However,
773 an impact-related origin seems the most plausible mechanism. In fact, LDG fragments are
774 thought to be the remains of a glassy surface layer, resulting from high temperature melting of
775 sandstones/desert sand, caused either by a meteorite impact, or - to explain the absence of an
776 impact crater - by airburst ([Aboud 2009; Seebaugh & Strauss 1984](#)). The Rare Earth elements
777 (REE) pattern of LDG is typical of upper continental crust, but the analysis of desert sand and

778 sandstones found in the Libyan Desert Glass strewn-field do not perfectly match, thus no rock
779 precursors and crater could be identified (Storzer & Koeberl 1991). Evidence for an impact
780 origin include the presence of schlieren and flow textures, detectable amount of Ir,
781 lechatelierite, and baddeleyite and, more in general, high pressure–temperature phases (Barrat
782 *et al.* 1997; Storzer & Koeberl 1991; Swaenen *et al.* 2010). Moreover, the high concentration
783 of PGE, the presence of graphite-rich inclusions (ribbons), and reduced Fe species, are
784 additional evidence for an impact origin (Barrat *et al.* 1997; Giuli *et al.* 2003; Pratesi *et al.*
785 2002). Some specimens of LDG present marked layering, with brownish or grayish-bluish
786 streaks (in general, both are referred as dark streaks). The brownish streaks are enriched in
787 siderophile elements, Mg, Fe and Ni, and the Os isotopic data indicates the extraterrestrial
788 component. Observations by Transmission Electron Microscopy (TEM) (Pratesi *et al.* 2002)
789 revealed the occurrence of Al-, Fe- and Mg-enriched amorphous nano-spherules (80-100 nm)
790 in the LDG dark streaks, as a result of silicate–silicate liquid immiscibility. Similarly, also other
791 impact glasses from Zhamanshin crater (blue zhamanshinites; Kazakhstan) contain spherical
792 nano-inclusions (~100 nm) of a second glass enriched in Fe, Ca, Mg and P (Zolensky & Koeberl
793 1991).

794 Based on the amorphous and mineral phases occurring in the different LDG specimens,
795 the range of temperature and pressure have been constrained to $T > 1900$ K and pressure from
796 10 to > 30 GPa (Pratesi *et al.* 2002; Gomez-Nubla *et al.*, 2017; Cavoisie and Koeberl 2019).
797 However, while high pressure occurred, most probably during melting and ejection, the
798 quenching of the molten material happened quickly at atmospheric pressure (Greshake *et al.*
799 2018). Moreover, the occurrence of meteoritic component suggests the impact of an
800 extraterrestrial object and not an airburst. This seems to be confirmed by a recent work by
801 Koeberl & Ferrière 2019 that reports the discovery of shock markers (*e.g.* PFs and PDFs on

802 quartz grains) in bedrock samples recovered in the Libyan Desert Glass strewn-field, and the
803 suggestion of a deeply eroded impact crater in the area.

804



805

806 **Figure 7 - 1) LDG with the typical aeolian erosion aspect 2) Slice of Darwin glass shows pronounced**
807 **layering and flow structures, and elongated vesicles.**
808

809

810 Another impact glass that cannot be referred to as tektite is found in a strewn-field of ~
811 400 km² in western Tasmania (Australia): the Darwin glass. The age of this glass, estimated by
812 Ar-Ar methods, is 816±7 ka (Lo *et al.* 2002), thus very close to the Australasian tektites.
813 Though, their major and trace elements compositions are very different, indicating that the
814 temporal association is casual. The Darwin crater, a small (~1.2 km) simple impact crater
815 formed in sedimentary target rocks, was discovered in 1972, and proposed as the source of
816 Darwin glasses (Ford 1972; Fudali & Ford 1979). Howard & Haines (2007) carried out a
817 detailed petrographic study of the crater-filling samples, but no conclusive shock markers have
818 been found (*e.g.*, PDFs in quartz grains). Nevertheless, the geochemistry of target rocks and
819 glasses, the location of the crater, and the glass distribution (Darwin strewn-field) all point to
820 the Darwin crater as source of the ejecta. Darwin glasses occur in an area larger than 400 km²,
821 with distances of at least 20 km (Howard 2011) from the putative Darwin crater, thus belonging
822 to the distal ejecta group. Glasses generally occur as irregular centimeter-sized fragments, or

823 masses, even if small splash-forms (spheres and teardrops < 5 mm) can be found across the
824 Darwin strewn-field. The color ranges from white, gray, light or dark green, dark brown, to
825 black, and the glasses are generally vesicular and often exhibit flow structure marked by bands
826 of elliptical vesicles (see **Figures 7 and 8**) (Glass & Simonson 2013). Interesting, the
827 proportion of white glasses is greatest in the proximity of the crater, and the proportion of darker
828 glasses, as well as the proportion of splashform shapes, increase with distance from the putative
829 crater (Gomez-Nubla *et al.* 2015; Howard 2008, 2009). Darwin glasses are SiO₂ rich, and
830 extremely depleted in Na₂O and CaO. The chemistry suggests the presence of (at least) two
831 main glass groups: the first one, which represents ~80% of the samples, is enriched in SiO₂
832 (average ~ 87 wt.%), whereas the second one has a lower SiO₂ content (average ~ 81 wt.%),
833 higher contents of MgO and FeO, and is enriched in Ni, Co and Cr (Howard 2008) (see **Table**
834 **2**). Optical microscope images of a Darwin glass show the contrast between layers of a lighter
835 coloured glass (light brown) and a darker one (dark brown); furthermore, turbulent flow
836 structures and bubbles of different dimensions and shapes are present (**Figure 8**). All textural
837 features visible in this specimen are very similar to those observed by See *et al.* (See *et al.* 1998)
838 in an impact glass bomb from the Ries crater. Raman spectroscopy data associated to the glass
839 specimen shown in **Figures 7 and 8** are reported in **section 4**. Analyses by Raman spectroscopy
840 allowed to identify small inclusions of α -cristobalite, and iron or iron/nickel oxides (Gomez-
841 Nubla *et al.* 2015).

842

843 Recently, Raman spectroscopy has been also employed to detect and identify the
844 organic matter in both LDG and Darwin glasses (Gómez-Nubla *et al.* 2018). Based on the
845 results, and the identification of *e.g.* fullerene-type compounds, the authors constrained a
846 pressure of ~15 GPa and temperatures between 670 and 1900 K. The preservation of organic
847 material in inclusions of LDG and Darwin glasses has been explained by the trapping of the

848 organic matter within the impact melt, and a fast quench that avoided the total decomposition
849 of the material (Gómez-Nubla *et al.* 2018; Howard *et al.* 2013).

850



851 **Figure 8 -** Optical microscope view (transmitted light) of the Darwin glass slice shown in
852 **Figure 7.** Darker and lighter layering, flow structures, vesicles and voids are clearly
853 **visible.**
854
855

856

857 **Formation of distal ejecta**

858 The origin and the formation mechanisms of tektites, microtektites and spherules have
859 been greatly debated. The terrestrial origin of tektites has been finally accepted, even if the
860 strong dispute started in the 50s (*e.g.* Barnes 1958; O'Keefe & Barnes 1958; Urey 1955, 1963)
861 has lasted until recently, with O'Keefe (1994) still supporting the theory that tektites were
862 ejected from volcanoes on the Moon (especially for the unanswered question related to the very
863 low water content of tektites), despite all geochemical and isotopic data available and the

864 provocative obituary by [Schnetzler \(1970\)](#) and his famous statement “The lunar origin of
865 tektites [...] died on July 20, 1969. The cause of death has been diagnosed as a massive
866 overdose of lunar data”. If the origin has been settled, the formation mechanisms are not
867 completely clear yet. New distal ejecta finding, improved experimental data and numerical
868 modelling have strongly enhanced the understanding of the physical-chemical conditions
869 occurring during a hypervelocity impact ([Melosh 1989, 2013](#)). Nevertheless, the reason(s) why
870 only a few impacts have produced significant amounts of distal ejecta is uncertain yet.
871 Moreover, the spherule formation from the plume is not fully constrained as well, because
872 physical and mathematical models are hampered by the limited information available on how
873 materials respond to extremely high stresses in highly non-equilibrium conditions. One of the
874 most important factors for distal ejecta formation is thought to be the obliquity of the impact
875 (the angle between the projectile and target impact), which determine the maximum shock
876 pressure and temperature regimes, and lately, the presence of water in the target rocks has been
877 pointed out as a key factor as well ([Artemieva 2002; Howard 2011; Stöffler et al. 2002](#)).

878 Distal ejecta are believed to be formed during the very initial stage of the excavation
879 (cratering process) when the projectile and the target materials (with a volume comparable to
880 that of the impactor) are highly shocked, melted and vaporized. The highly pressurized mixture
881 of vaporized rocks, molten materials, small solid fragments, and hot atmospheric gases
882 constitute the so-called vapor plume (aka impact plume or fireball) ([Melosh 2013](#)), that expands
883 away from the forming crater. The vapor plume pushes away the ambient atmosphere in its path
884 and this allows ejected melt to follow long ballistic trajectories (*cf.* [Howard 2011](#); see also
885 [Melosh 1989](#)), and for large enough impacts the vapor plume might expand and burst the top
886 of the atmosphere allowing materials to be dispersed at great distances, even at the global scale.
887 Numerical modeling on spherules (or tektites) nucleation and condensation from the vapor
888 plume has been done by *e.g.* [Johnson & Melosh 2012, 2014; Melosh 1989; Pierazzo et al. 1998](#).

889 Johnson & Melosh 2012, 2014 differentiate two origins for spherules, based on their
890 thermodynamic histories: i) vapor condensed droplets and ii) melt droplets, in agreement with
891 early observations on other ejecta (Engelhardt *et al.* 1987). Because of the extremely high
892 temperatures expected in the impact plume, the melt will have an extremely low viscosity ($7 \times$
893 10^{-6} Pa s, according to Melosh & Artemieva 2004), and volatile-bearing bubbles will rapidly
894 travel through the melt and burst at the liquid surface, resulting in a liquid phase quickly
895 depleted in water and other volatile elements, such as Na and K (Melosh & Artemieva 2004).
896 The presence of a superheated melt in the vapor plum might explain not only the occurrence of
897 alkali depletion, and the low water content in the ejecta, but also the oxygen volatilization and,
898 in turn, the highly reducing conditions.

899

900 3.1.2 Proximal impactite

901 Glasses deposited within five crater radii from the impact site are referred to as proximal.
902 These amorphous materials can resemble tektites, but they show higher chemical heterogeneity,
903 relatively higher water contents (0.02 to 0.06 wt.% H₂O), and contaminations from the impactor
904 (Devouard *et al.* 2014; Koeberl *et al.* 2019; McPherson *et al.* 1984). Example of glasses
905 associated with impact sites are Irghizites (Kazakhstan), Aouelloul glasses (Mauritania), Lonar
906 Crater Glass (India), Waber Glass (Saudi Arabia) and Atacamaites (Chile).

907 Associated to the Zhamanshin impact structure (Kazakhstan) there are a variety of impact
908 glasses. The main groups are i) Irghizites, glasses with aerodynamic flight-shapes that resemble
909 tektites, but with higher water contents, and with an average SiO₂ content of ~74 wt.%; ii)
910 Zhamanshinites that can be found closer to the source crater and are much larger than Irghizites.
911 Zhamanshinites are very heterogeneous, with layered structures, flow structures and based on
912 the chemistry they are further differentiated in different subgroups (Fredriksson *et al.* 1977;
913 Koeber & Fredriksson 1986), with glasses Si-enriched (> 70 wt.% SiO₂), and glasses strongly

914 depleted in silica (< 55 wt.%), called basic impactites. An additional amorphous material is
915 referred to as blue Zhamanshinite. It has high SiO₂ and CaO contents, and present evidences of
916 liquid immiscibility with the occurrence of amorphous inclusions that provide the characteristic
917 turquoise to very dark blue color (Koeber & Fredriksson 1986; Koeberl 1988; Zolensky &
918 Koeberl 1991; Koeberl 1988; Zolensky & Koeberl 1991). Indeed, the nano-sized Ca-Fe-Mg-P-
919 rich silicate immiscible liquid inclusions are responsible of the scatter of light (Rayleigh
920 scattering) that results in the blue coloring of the glass.

921 Africa has 19 confirmed impact structures (Reimold & Koeberl 2014), and among them
922 there is the Aouelloul structure in Mauritania, a relatively young (3.1 ± 0.3 Ma) crater of ~390
923 m diameter. The enrichment in siderophile elements, and the occurrence of lechatelierite,
924 baddeleyite, Ni-rich Fe- spherules are the impact marker identified. The glasses associated
925 (Aouelloul glasses) spread in proximity of the crater, and similarly to the Darwin glasses, are
926 very heterogeneous. They are water-depleted silicate glasses, with abundant schlieren texture
927 and different bulk chemistries, and with the presence of unambiguous meteoritic component.

928 Wabar crater is a very young impact structure in Saudi Arabia (290 ± 38 years). The
929 associated glasses are both large fragments and small (≤ 1 cm) glassy aerodynamic flight-shaped
930 and spheres [Prescott *et al.* 2004; Hamann *et al.* 2013]. Wabar glasses are known to have several
931 chemical heterogeneities, with flow structures, emulsions of Fe-rich ultrabasic silicate glasses,
932 FeNi globules and areas marked with light- and dark-brown glasses, respectively depleted or
933 enriched in Fe (Hamann *et al.* 2013; Hörz *et al.* 1989; Ottemann 1966; see Table 2). See *et al.*
934 (1998) by studying the glasses microstructure, highlighted the textural and chemical
935 heterogeneity that are present in these impact glasses, and the occurrence of interstitial
936 enrichments of Fe and Ni, most probably resulting from the impactor.

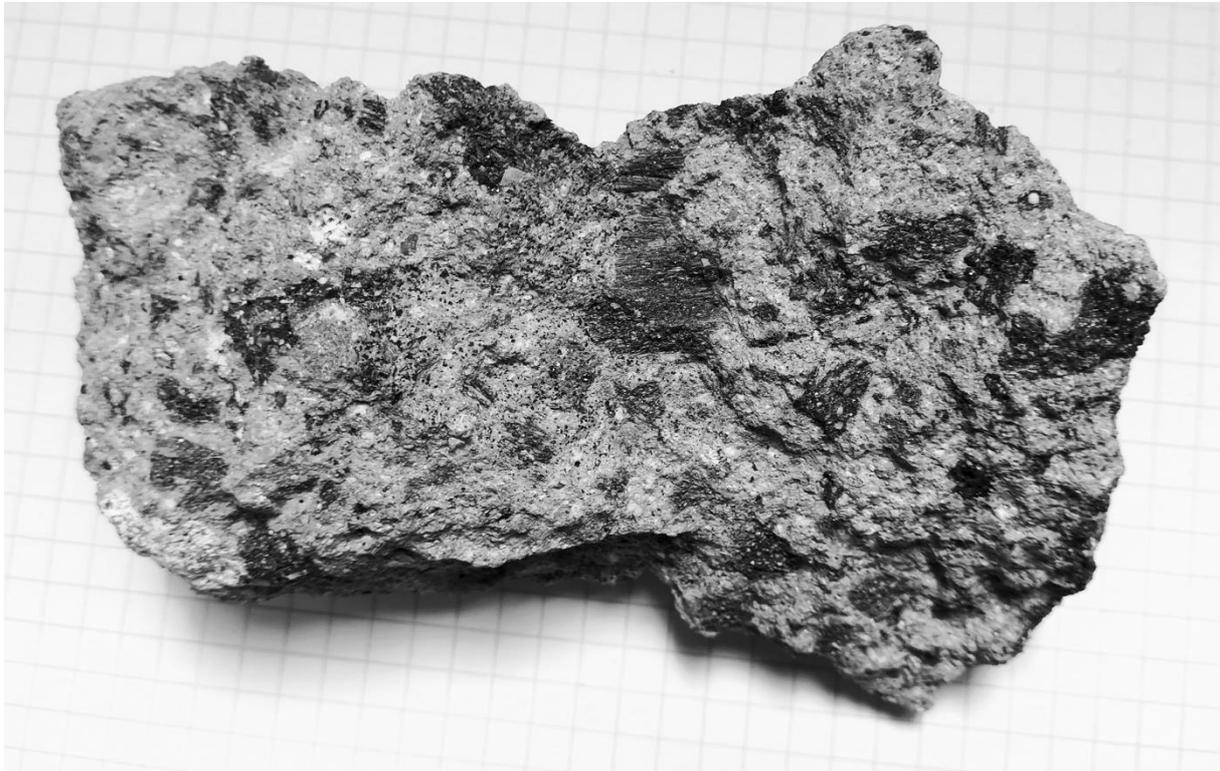
937 From Atacama Desert (Chile) thousands of aerodynamically shaped black glasses have
938 been recovered and named atacamaites. These glasses have minor vesicularity, inclusions of an

939 extremely silica rich glass (lechatelierite), turbulent flow structures, strong chemical variations
940 (e.g. FeO contents vary from 5 wt.% to 15 wt.%), and extremely high contents of Ni, Co, Re,
941 Ir and Pt (orders of magnitude higher than the average upper continental crust). Moreover, water
942 contents are higher (~ 130 ppm) than those associated to tektites. All these characteristics point
943 to the impact nature of these glasses (Devouard *et al.* 2014; Dos Santos *et al.* 2015; Koeberl *et*
944 *al.* 2019).

945
946 For sake of comprehensiveness, we will briefly describe an example of a proximal
947 impact breccia, the suevite, from the Ries crater type locality. An exhaustive review on Ries
948 crater and suevite is reported by Stöffler *et al.* (Artemieva *et al.* 2013) (the modelling part is in
949 Artemieva *et al.* 2013). The term suevite was firstly used to describe an impact breccia
950 formed at the Ries impact crater (Bavaria, Germany); nowadays the term is used more in general
951 to describe polymict (consisting of heterogeneous fragments) impact breccia containing clastic
952 matrix/groundmass from the target rocks, shocked minerals, and glass. At the Ries impact site,
953 suevite can be found both in the crater and outside the rim (outer or fallout suevite), associated
954 to the different (five) shock stages. Because of the occurrence of a full set of rocks subjected to
955 different degrees of shock metamorphism (and distal ejecta as well), the Ries crater is one of
956 the most studied, and has been the type locality in order to discover and classify all shocked
957 materials (Artemieva *et al.* 2013; Stöffler 1971). Figure 9 reports a photo of a glass-rich suevitic
958 impact breccia (fallout suevite from Otting quarry; Ries crater, Germany). The heterogeneous
959 nature of the sample can be appreciated, with lithics, relict mineral grains and abundant glass
960 portions. From this locality it was first reported the occurrence of coesite (Shoemaker & Chao
961 1961). According to Stöffler *et al.* (Artemieva *et al.* 2013 and references therein) the fallout
962 suevite (also called outer suevite) is almost completely constituted of lithic clasts derived from
963 the target crystalline basement rocks, and only a few percent is of sedimentary origin (< 6%
964 sedimentary lithic) and contain rocks of all stages of shock metamorphism, from the unshocked

965 materials of stage 0 (0-10 GPa), to moderate shocked (stages I and II: 10-60 GPa), up to stage
966 IV (> 60GPa) and there is the occurrence of abundant glass materials.

967



968

969 **Figure 9 - Fallout suevite from Otting quarry (Ries crater, Germany). The polymictic sample**
970 **has lithics (brighter spots), relict mineral grains and abundant glass (darker portions).**

971

972 [Oberdorfer \(1905\)](#), in suevite deposits in the west side of the Ries crater, observed the
973 presence of spherical dark-brown amorphous bodies surrounded by a light brown glass, with
974 textural features indicative of silicate-silicate melt immiscibility. Other textural evidences of
975 liquid immiscibility were reported by [Graup \(1999\)](#), but in this case for an immiscibility
976 between silicate and carbonate melts. Crystalline calcite is a common constituent of suevite,
977 and it was believed to derive from post-impact hydrothermal activities ([Engelhardt 1967, 1972](#)).
978 However, because of the presence of gas vesicles in carbonate flow lines, or carbonate globules
979 embedded in silicate glass (and *vice versa*), and the presence of deformed and coalescing

980 spheres of carbonate within the silicate melt, the carbonate component is likely a result of
 981 impact-shock melting, and not of secondary origin (Graup 1999).

982

983 As observed above, impact glasses (including the enigmatic ones) are more
 984 heterogeneous than distal ejecta (*i.e.* tektites). For example, Irghizites, Wabar and Darwin
 985 glasses clearly present at least two major chemical bulk compositions. In Table 2 is reported
 986 the average major oxide composition for some of the impact glasses discussed here. The
 987 chemical variability among the same impact glasses, and with respect tektites (see data in Table
 988 1) reflects the differences in the mechanisms of formation of those glasses, especially related
 989 to the different time-scales.

990

991 **Table 2: Average major oxide compositions of impact glasses (wt.%). Data from § Koeberl 1997;**
 992 **⊕ Giuli *et al.* 2003; # Ottemann 1966; ‡ Howard 2008; \$ unpublished data.**

993

	LDG §	LDG dark streak ⊕	Wabar dark #	Wabar light #	Aouelloul #	Suevite glass - Ries #	Darwin ‡	Darwin* \$	Atakamaite \$
SiO₂	98.4	95.85	87.45	92.88	86.51	66.19	76.47 - 93.85	82.74	68.74
TiO₂	0.12	0.18	0.15	0.12	0.55	0.82	0.22 - 0.8	0.609	0.43
Al₂O₃	1.19	1.48	1.77	2.64	5.76	14.74	3.14 - 11.45	7.89	10.66
FeO	0.12	0.98	5.77	0.53	1.59	3.86	0.84 - 5.87	2.39	5.40
MnO		0.02						0.01	0.06
CaO	0.01	0.08	1.90	1.46	0.73	3.96	0.02 - 0.25	0.04	3.59
MgO	0.01	1.38	0.60	0.47	0.91	2.60	0.24 - 4.0	0.87	1.52
Na₂O	0.005	0.02	0.39	0.42	0.14	3.62	0.0 - 0.21	0.09	2.77
K₂O	0.009	0.01	0.58	1.61	2.05	3.26	0.75 - 2.71	2.05	2.61

994 Note: * average chemical composition of the specimen shown in Figure 8

995

996

997 **3.2 Fulgurite**

998 Fulgurites are formed as a result of fusion and modifications of rocks (*s.l.*) by lightning,
999 and the name derives from the Latin word “*fulgur*” (lightning). Usually they consist of
1000 irregularly shaped tubes ranging from approximately 1 cm in diameter to 1 mm, even if they
1001 may extend laterally or vertically for up to 10 m (Essene & Fisher 1986). Famous fulgurites
1002 specimens are those coming from the Saharan desert; however, fulgurites can occur on any soil,
1003 either formed naturally or triggered in proximity of high voltage power lines.

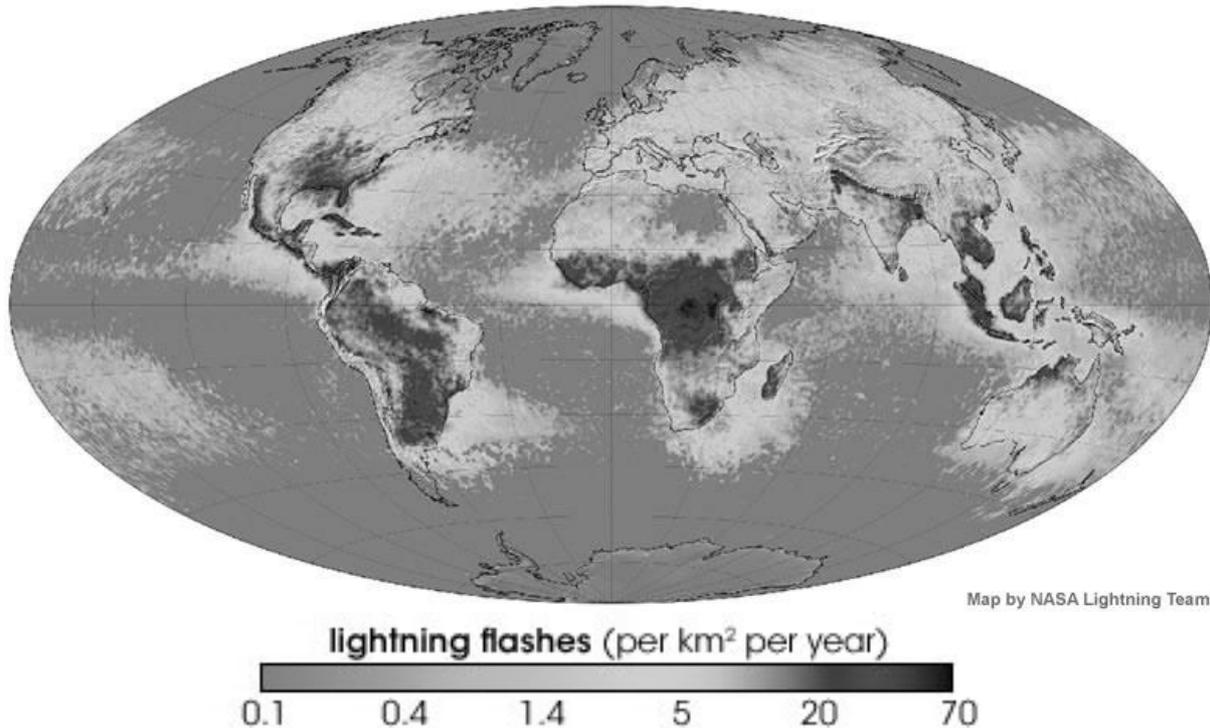
1004 The study of the mineralogy and petrology of these specimens, and of their mechanisms
1005 of formation has started to become more and more significant. Starting from the curiosity of
1006 understanding more in detail these fascinating objects, researcher from physics, atmospheric
1007 sciences, geochemistry, materials science, impact-related sciences have gathered and compared
1008 many information, and the results are well beyond the pure scientific curiosity, with key
1009 implications in biogeochemistry and formation of life. In this section we will try to introduce
1010 the lightning process, the fulgurite morphology and mineralogy, and we will try to arouse
1011 interest on the multidisciplinary aspects related to fulgurites.

1012

1013 Lightning is a very common phenomenon but is not uniformly distributed across the Earth
1014 (see image from Lightning Imaging Sensor (LIS) NASA... Ask for permission!) and equatorial
1015 conditions (warm, moisture!) greatly favor the occurrence of lightning. As a curiosity, the place
1016 on Earth with the highest lightning flashes for year is Lake Maracaibo (Venezuela) with an
1017 average of ~232 flashes (per km² per year).

1018

1019



1020

1021

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1025

Figure 10: World Lightning Map shows the average yearly counts of lightning flashes per square kilometer based on data collected by NASA's LIS between 1995 and 2002. Localities with the largest number of lightning strikes are in the Equatorial area.

1026

1027

This common phenomenon is extremely energetic, and lightning may dissipate $\sim 10^9$ J

1028

([Borucki & Chameides 1984](#); [Rakov & Uman 2003](#)), raising, instantaneously (< milliseconds),

1029

the surrounding air temperatures in the range of 10^5 K ([Uman 1964](#); [Uman & Krider 1989](#)).

1030

Thus, a really high amount of energy strikes the surface target material, providing a local

1031

temperature rise higher than 2000 K, and with even greater temperatures expected on the

1032

“current” path ([Uman & Krider 1989](#)). Such amount of energy is able to melt/vaporize the

1033

inorganic target materials, and incinerating the organic ones along the current path, inducing

1034

the formation of voids and vesicles from the release of the volatilized components. The heating

1035

rates have been constrained to be in the order of 10^3 K/s ([Carter et al. 2010](#); [Jones et al. 2005](#);

1036

[Krider et al. 1968](#); [Uman 1964](#)), whereas the cooling rates are estimated in the order of 10^8 K/s

1037

([Rietmeijer et al. 1999](#); [Switzer & Melson 1972](#)) because of the formation of very homogeneous

1038

glass layers and the abundance occurrence of lechatelierite. The result of such fast event is a

1039 cylinder (often presenting lateral branches) with an inner smooth glassy portion surrounded by
1040 an irregular outer surface that can be composed of melted, partially melted and unmelted
1041 minerals grains and rock fragments, and with many spherical and elongated vesicles occurring
1042 in all portions (**Figure 11**).



1043

1044 **Figure 11 –Fulgurite specimens showing different morphologies. TO CHANGE???**

1045

1046 Lightning strikes are, therefore, rapid high energetic phenomena able to induce strong
1047 physical and chemical modifications in the target materials, and the morphology of the fulgurite
1048 specimens formed will be strongly dependent on energy and timescale of the striking, on target
1049 composition and heterogeneity, and so on. The bulk composition of fulgurites is quite broad
1050 when considering the whole samples, as a reflection of the different target materials (**Figure**
1051 **1**). However, the chemistry of the glassy part is much narrower and reveals a volatilization of
1052 most of the major elements, resulting in an amorphous phase enriched in silica, and depleted in
1053 alkalis. Most of the studies report the occurrence of glass heterogeneities, with lechatelierite
1054 glass (98-100 wt.% SiO₂) occurring in different percentages, and secondary glass phases having
1055 relatively lower amounts of SiO₂, and higher proportions of Al₂O₃ and FeO ([Essene & Fisher](#)

1056 [1986; Frenzel & Stahle 1984; Pasek *et al.* 2012](#)). [Pasek *et al.* \(2012\)](#) established a classification
1057 for fulgurites based on morphology, mineralogy and petrology, providing five main types
1058 depending on the target rocks (sand, soil, rocks):

1059 • Fulgurites formed in quartz sand are classified as type I, and usually present thin glassy
1060 walls; type I can contain one or two melts consisting prevalently of lechatelierite, and
1061 sometimes, also a SiO₂-rich melt with higher concentrations of Al and/or Fe. In the groundmass
1062 has been reported also enrichments in Zr oxide- and Fe–Ti oxide- rich glass.

1063 • Fulgurites type II has a thicker glass portion compared to type I. The amorphous portion
1064 is more compositionally varied (< 50% lechatelierite), because formed in different
1065 environments than silica sand, such as unconsolidated soils containing clays minerals, small
1066 rocks, and so on.

1067 • Type III consists of lechatelierite and feldspar glasses, and a calcite-rich matrix.
1068 Fulgurites type III are mostly found in calcite-rich soils and are the densest (average density¹
1069 $2.1 \pm 0.5 \text{ g/cm}^3$).

1070 • Type IV fulgurites are heterogeneous melts, with the inner portion rarely consisting of
1071 lechatelierite, and the outer portion consisting of unmelted (or partially melted) rocks and
1072 mineral grains. These fulgurites form usually in bulk rocks and have densities similar to those
1073 of the target rocks.

1074 • Type V are droplet fulgurites that have a homogeneous glass matrix. The two main
1075 oxides contained in type V droplets are enriched in SiO₂ and K₂O relative to the originating
1076 fulgurite, whereas other oxides are depleted.

1077 An updated classification ([Pasek & Pasek 2018](#)) includes also specimens artificially
1078 formed, either from high voltage power lines, or triggered by high-voltage electrical arcing.

¹ The density value reported is related to the density of the material as approximated by a whole fulgurite cylinder [Pasek *et al.* 2012](#)..

1079

1080 Since fulgurites result from a quick high energetic event that produces also a shock wave,
1081 in many fulgurite specimens shock markers typical of impact glasses have been identified. For
1082 instance, shock lamellae in quartz, kink bands in plagioclase have been reported, with estimated
1083 shock pressures >10 GPa [[Gieré et al. 2015](#)]. Other studies (either experimental or based on
1084 modeling) reported even higher shock peak pressure, above 20 GPa [[Carter et al. 2010](#); [Ende
1085 et al. 2012](#)]. Artificial produced (triggered) fulgurites confirmed that temperature and pressure
1086 regimes needed to produce shocked materials can be achieved by lightning strikes ([Chen et al.
1087 2017](#)). The occurrence of such shock features in fulgurites, until then considered diagnostic of
1088 impactites, raised questions about samples of uncertain origin, such as irregular glassy objects
1089 having shocked quartz grains, but unlikely related to any impact event ([Melosh 2017](#)).

1090 Besides the occurrence of shock markers, in many fulgurites have been observed O-free
1091 phases and metals indicative of extremely reducing conditions, such as those typical of
1092 meteorite phases. These findings have extremely important implications, especially concerning
1093 the origin of life (see below). The occurrence of metallic phases have been reported since long
1094 ([Anderson 1925](#); [Essene & Fisher 1986](#); [Wasserman & Melosh 2001](#)), and [Essene & Fisher
1095 1986](#), explained the occurrence of metallic globules rich in silicon, spheroids of silicon-bearing
1096 metals (99.5 at.% of metallic silicon phase with minor amounts of titanium, iron, and
1097 phosphorus), and Fe-rich spherules, via thermodynamic calculations that indicate that
1098 extremely high temperatures (> 2000 K) and reducing conditions close to the SiO₂-Si buffer
1099 were needed. However, the authors could not decipher (and rule out) the role of other agents,
1100 such as the presence of carbonaceous materials, or the degassing of oxygen or the formation of
1101 nitrogen oxide gases.

1102 Other metallic phases, such as Fe-Si-Al-Ti metallic droplets, or phosphide phases usually
1103 common in meteorites (*e.g.* Fe₂Si, Fe₃P) have been reported as well ([Pasek et al. 2012](#) and

1104 [references therein](#)]. [Rowan & Ahrens 1994](#) report phases reduction in MORB glasses by shock
1105 experiments. Shock pressures between ~0.8 and ~6 GPa induced a change of the glass chemistry
1106 (enrichment in SiO₂) and the formation of Fe-, Si- and Mo-rich metallic microspheres
1107 embedded in the shocked glass. Based on the experimental conditions and the occurrence of
1108 iron silicide alloys, the authors estimated that an oxygen fugacity ~ 8 log units below the initial
1109 one was needed in order to form such metallic phases. [Jones et al. \(2005\)](#) artificially (triggered-
1110 lightning) produced specimens composed of 99.9% pure binary oxides of manganese and nickel
1111 in order to study the reduction mechanisms, and while they observed the formation of nickel
1112 oxide particles, the manganese oxide fulgurite showed no metallic phase formation. Thus, the
1113 thermodynamical stability of the oxides involved has a key role in the reduction mechanisms.
1114 Based on their experiments, [Jones et al. \(2005\)](#) suggest different possible mechanisms for the
1115 reduction associated to lightning, and they highlighted that the presence of carbon is not
1116 required for oxide reduction during fulgurite formation.

1117 The presence of reduced phosphorous species in fulgurites is particularly important. A
1118 relative enrichment in phosphorous is not unusual in fulgurites ([Martin Crespo et al. 2009](#);
1119 [Pasek et al. 2012](#); [Pasek & Block 2009](#)), but the occurrence of phosphides has different
1120 implications. In iron meteorites, pallasites and enstatite chondrites, phosphorus is commonly
1121 occurring as phosphide (*e.g.* schreibersite (Fe,Ni)₃P), which slowly react with water to produce
1122 reduced forms (PO₃³⁻ phosphite). Contrarily, on Earth, phosphorus occurs in minerals with its
1123 stable form P⁵⁺ (PO₄³⁻ phosphate). Besides the mineral supergroup apatite (the primary source
1124 of P), this element is a common constituent of organic molecules (DNA, RNA, and so on...).
1125 On Earth surface, a possible mechanism for reduction of phosphate species may be lightning,
1126 as shown by simulated discharge experiments from [Glindemann et al. 1999](#), and from the
1127 studies on many fulgurite specimens by Pasek and coauthors. For example, [Pasek and Block](#)

1128 (2009) report that the reduction of PO_4^{3-} species to reduced forms occurs commonly in many
1129 target soils, and they provided two potential causes of P reduction during fulgurites formation:

1130 1) the reduction of phosphate to phosphide, and a later reaction with water; this process
1131 is analogous of that of meteoritic schreibersite that react with water to produce reduced forms
1132 e.g. H-phosphonic acid H_3PO_3 (e.g. Bryant *et al.* 2009);

1133 2) the direct reduction from PO_4^{3-} to PO_3^{3-} species; thermodynamical calculations indicate
1134 that phosphates (CaHPO_4) could be reduced to phosphite species (CaHPO_3) at temperatures
1135 ~ 2000 K, under oxygen fugacity conditions of ~ -5 (as $\log(f\text{O}_2)$) (Pasek & Block 2009).

1136

1137 The phosphides species occurring during meteoritic impacts, and especially during the
1138 Hadean-Archean heavy meteorite bombardment have been proposed as a way of altering global
1139 phosphorus biogeochemical cycle, and the source of reactive phosphorus required for the
1140 processes of prebiotic phosphorylation on the early Earth (Pasek 2008, 2017; Pasek & Block
1141 2009).

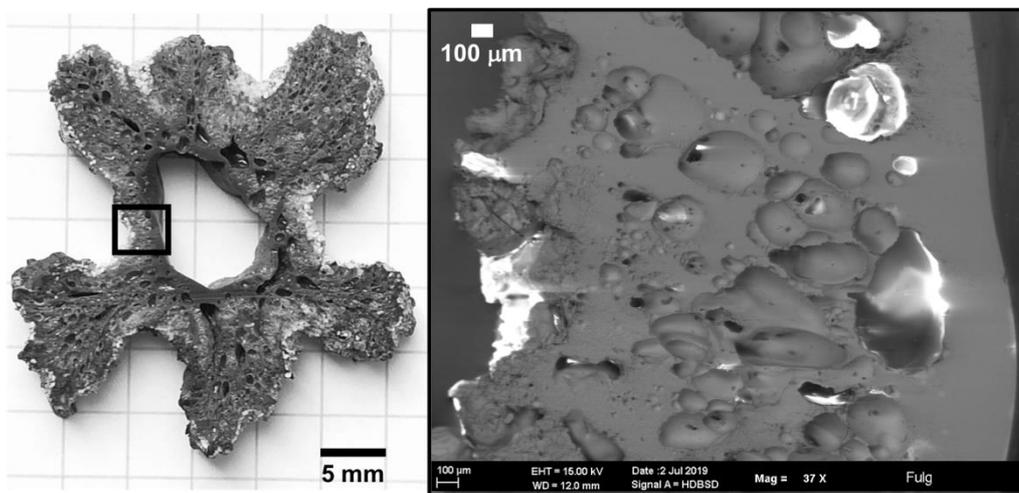
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1143

1144 **Case study: fulgurite**

1145 A fulgurite with a dark thick glassy inner core has been used for a series of analysis. The
1146 specimen is highly vesiculated and the inner glass core has a variable thickness up to 1 mm.
1147 The outer portion presents unmelted white/grey grains of quartz, and probably salts species.
1148 The specimen has been sliced and the cross section polished (**Figure 12**). Optical microscope
1149 images, and Raman spectroscopy analysis have been coupled to SEM/EDS data in order to
1150 provide an overview of all the phases occurring in this fulgurite.

1151 The polished slice has been observed with an optical microscope by reflected light, and
1152 SEM. The glass matrix of the inner part seems to be very homogeneous, with no relict mineral
1153 grains detectable, and a few bubbles if any. Moving from the inner portion toward the external
1154 wall (see **Figure 12**), vesicles, mostly elongated, are clearly increasing in amount, and the matrix
1155 become more heterogeneous. The fulgurite specimen has several outer branches, and many
1156 large voids.



1157
1158 **Figure 12:** Photograph of the sliced fulgurite (left), and SEM image (right) of the thinner portion where
1159 both inner and outer walls can be observed. The inner wall has a smoother surface, in contrast with the
1160 rough outer part. Brighter areas in the left image are residual of carbon coating.
1161

1162 Backscattered electron images of the fulgurite (**Figure 13**) show the increasing number of
1163 elongated bubbles, which radially distribute in the branches. The matrix is predominantly
1164 amorphous and fairly homogeneous close to the inner wall, while all outer branches are more

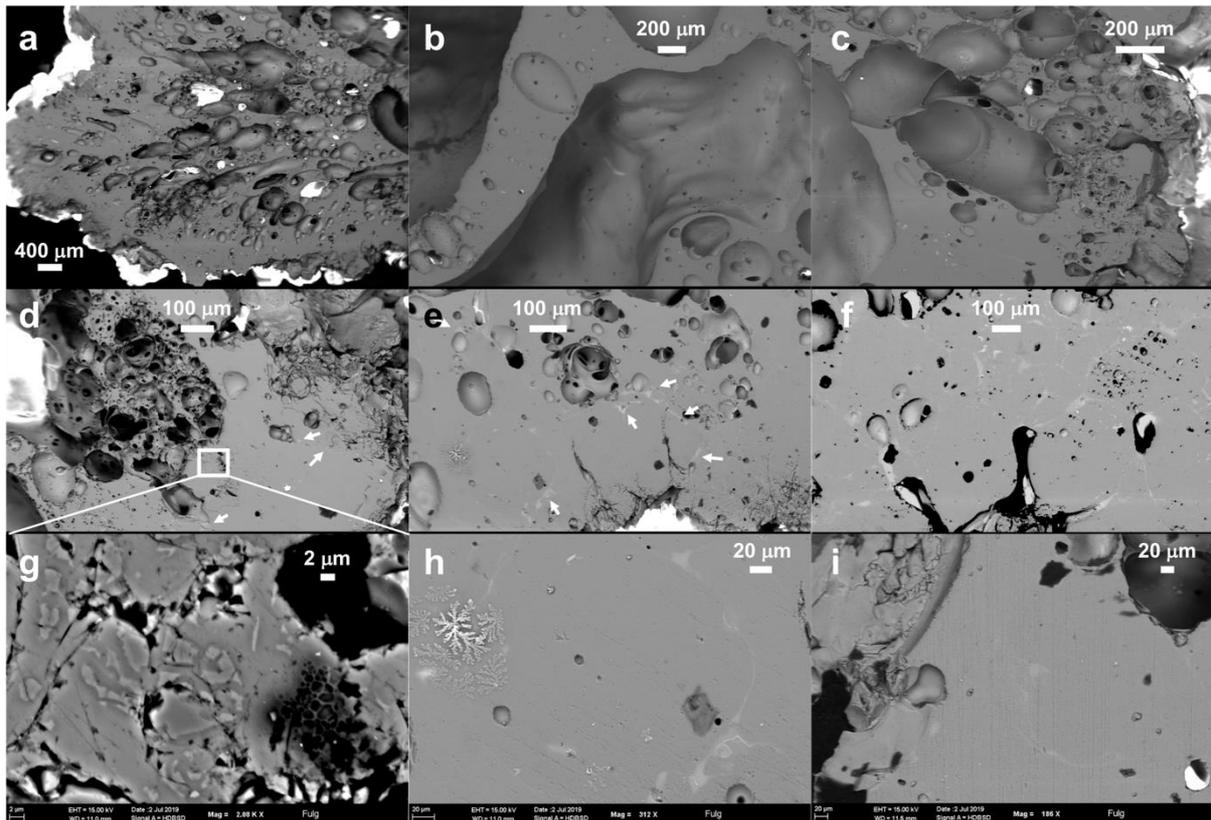
1165 heterogeneous, with completely foamed glass portions, unmelted quartz grains, regions with
1166 small zoned crystals, and tiny bright spots, too small to be characterized.

1167 EDS analyses of the inner glass portion reveals the homogeneous presence of
1168 lechatelierite, with small amounts of Al_2O_3 ($\text{SiO}_2 \geq 99$ wt.%, $\text{Al}_2\text{O}_3 \leq 1$ wt.%). When moving
1169 from the smooth inner to the rough outer walls the chemical compositions marginally differ,
1170 but without a defined trend. The SiO_2 content ranges between 96.6 and 99.5 wt.% with some
1171 random points having higher Al_2O_3 amounts (~ 2.4 wt.%) and traces of FeO, MgO, CaO and
1172 K_2O (all < 0.4 wt.%).

1173 The areas where the branches depart and all the branches themselves have areas (pockets)
1174 and veins-like features that especially surround bubbles (see arrows in **Figure 13**). These
1175 portions are highlighted in the BSE images by the different contrast (light grey) and the EDS
1176 analysis indicate lower Si contents (down to ~75 wt.%), and higher amounts of Al_2O_3 (up to ~
1177 14 wt.%), and the presence of many other elements, including FeO (3-6.5 wt.%), MgO (1-1.5
1178 wt.%), K_2O (0.5-1 wt.%), CaO (~ 0.4 wt.%) and TiO_2 (~ 0.3 wt.%). The elemental mapping of
1179 one of the regions rich in pockets and veins is reported in **Figure 14**. The primary matrix is
1180 composed almost exclusively by SiO_2 , and only a 0.5-1.5 wt.% of Al_2O_3 . The bubbles are
1181 surrounded by large light grey pockets and veins that contain the secondary melt relatively
1182 depleted in silica, and enriched in Al, Fe, and alkali/alkali-earth elements. In a few light grey
1183 spots there are also traces of P_2O_5 and Na_2O (~ 0.3 wt.%).

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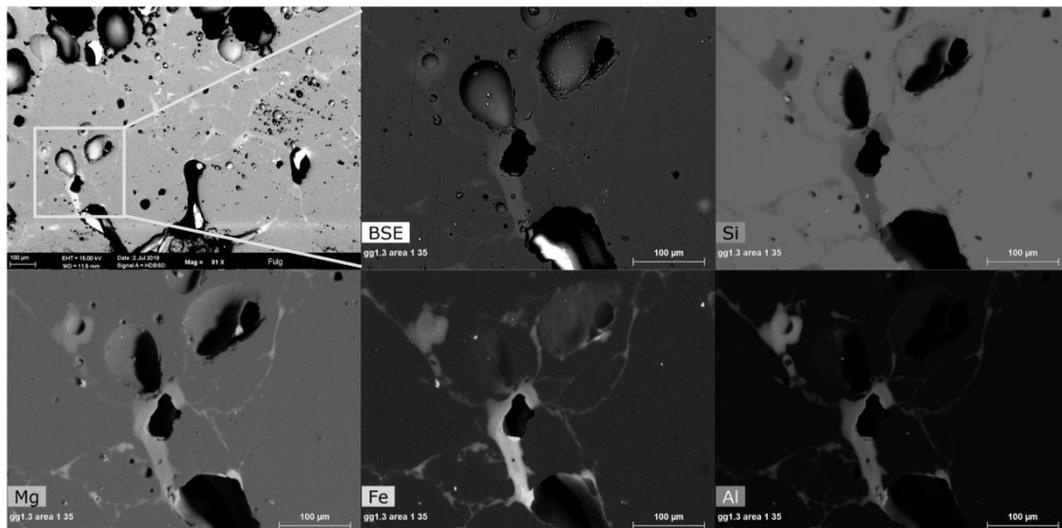
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Figure 13: backscatter electron images of the fulgurite at different magnifications to emphasize the heterogeneity of the sample. Overview of the a) outer branch, b) inner glass wall, c) outer portion in the thinner area; d-e-f) outer portions with heterogeneous features, such as foamed area, aligned bubbles, relict quartz grains and pocket and veins of glass with different chemistry (light grey; arrows); g-h-i) higher magnifications.



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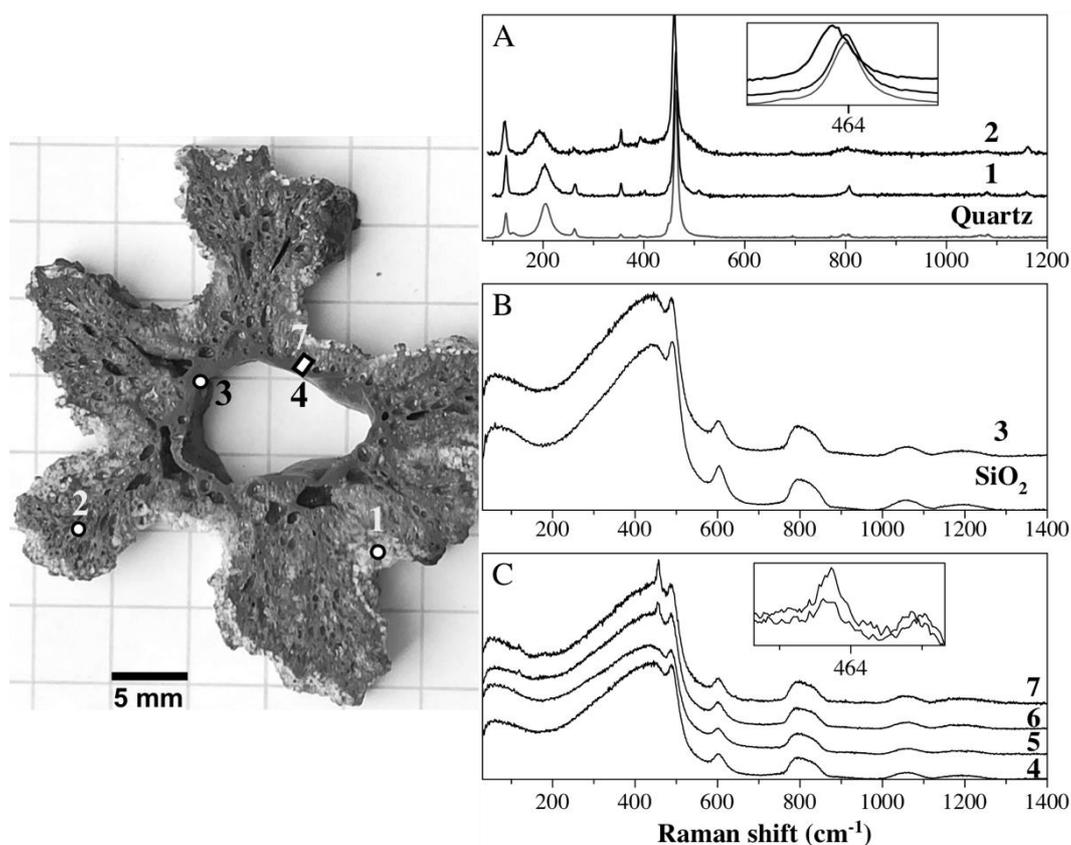
Figure 14: Elemental mapping of one of the regions rich in pockets and veins. The primary glass matrix is composed of ~99 wt.% SiO₂, and voids and bubbles are surrounded by a secondary melt, relatively depleted in Si, and enriched in Al, Mg and Fe. Scale bar in all images is 100 μm.

1199

1200 Raman spectra were collected in several portions of the sliced fulgurite, with a 488 nm
1201 excitation laser, in order to characterize the amorphous and the mineral phases. The outer whiter
1202 grains, supposedly residual of the target rocks, are mainly composed of quartz and some salts,
1203 such as NaCl (from EDS point analyses). The spectra of the outer grains perfectly match the
1204 spectra of α -quartz (see [Figure 15A](#)), with the presence of a sharp intense peak at $\sim 464\text{ cm}^{-1}$,
1205 and a broad band at $\sim 205\text{ cm}^{-1}$ (A_1 modes), and also the E modes at $\sim 127\text{ cm}^{-1}$ and 263 cm^{-1} .
1206 The differences in the relative intensity of the E modes between the reference quartz spectrum
1207 and the spectrum of the outer grains (point 1 in [Figure 15A](#)) is due to sample orientation. Similar
1208 spectra were obtained in some of the branches, although with some of the main peaks shifted
1209 (point 2 in [Figure 15A](#)). Indeed, all peaks match vibrations of alpha-quartz, but with shifts of
1210 the vibrational modes that indicate the occurrence of shock pressures ([Table 3](#)). For instance,
1211 while the main peak moves $\sim 3.1\text{ cm}^{-1}$ toward lower frequencies, the broad peak at 205 cm^{-1}
1212 shifts more than 10 cm^{-1} downward, and the small peak at 354 cm^{-1} remains unchanged. These
1213 uneven shifts of the quartz peaks indicate the presence of shocked quartz, and based on the
1214 experimental work done by [McMillan et al. \(1992\)](#), the shifts observed provide a shock peak
1215 pressure between 25 and 28 GPa, similarly to what reported by [Carter et al. \(2010\)](#).

1216 The Raman spectrum of the glass on the inner wall (point 3 in [Figure 15B](#)) has the clear
1217 broad features of amorphous silica and nearly perfectly match the spectrum of SiO_2 suprasil,
1218 used here as reference (SiO_2 in [Figure 15B](#)). These spectra agree with the SEM/EDS analysis
1219 and the occurrence of a homogeneous lechatelierite glass in the inner wall. When moving from
1220 the inner wall toward the outer one (points 4 to 7 in [Figure 15C](#)), the shape of the Raman spectra
1221 is very similar, and small variations can be attributed to the different concentration of Al_2O_3 in
1222 the amorphous matrix. Moreover, the two last spectra show the presence of small sharp peaks,

1223 that indicate the occurrence of microcrystalline quartz. Based on the position of the main peaks
 1224 here the shifts suggest higher shock pressure, of ~ 30-31 GPa (Table 3).



1225
 1226 **Figure 15:** Raman spectra of different portions of the fulgurite (numbers from 1 to 7) and crystalline
 1227 references. The outer unmelted grain and a small grain in the branch present vibrations associated to
 1228 alpha-quartz (2). The glass on the inner wall (3) is very polymerized and the spectra is very similar to that
 1229 of a pure SiO₂ glass. Moving from the inner wall toward the outer part (from 4 to 7) the homogeneous
 1230 glass matrix start to be slightly more heterogeneous with the presence of a small amount of quartz (sharp
 1231 peaks in 6 and 7 and inset).
 1232

1233 **Table 3:** Frequencies of the main vibration modes of α -quartz (reference phase), and of the
 1234 crystalline phases occurring in the fulgurite specimen. Numbers refer to the sampling spots
 1235 reported in Figure 15.

Reference	Spectrum 1	Spectrum 2	Spectrum 6	Spectrum 7
α -quartz				
126.8	126.8 cm ⁻¹	124.1	120	120
205.1	204.5	194.1		
263.1	263.6	262.1		
354.2	354.2	354.2		
463.6	463.6	460.5	457.8	457.8

1236

1237

1238 **3.3 – Trinitite or nuclear glass**

1239 Within glasses formed by highly energetic events, there is a glass that is not natural, but
1240 neither man-made in the strict definitions. During the first atomic bomb test in Alamogordo
1241 (USA Trinity test, 1945), the detonation of the nuclear weapon on the desert sand has created a
1242 *“Lake of Jade [...] a lake of glistening incrustation of blue-green glass 2400 feet in diameter”*
1243 as reported by Time Magazine (Science: Atomic footprint, 17 Sept 1945). The green amorphous
1244 material has been called trinitite, or nuclear glass. Another nuclear test that produced
1245 amorphous material is the first underground nuclear explosion, the so-called Rainier test (1957).

1246 The study of these glasses has mainly focused on the radioactivity of the materials, and
1247 on the incorporation and distribution of radionuclides, although, more recently, more detailed
1248 chemical, mineralogical and geochemical investigations have been reported. Moreover,
1249 because of the mechanisms of formation, trinitite could be regarded as an analogous of
1250 impactites.

1251 In the following, we are going to discuss about the event associated to the formation of
1252 trinitite, and we will provide information on the different geochemical aspects of this glass.



1253 **Figure 16. Photograph of a bottle green trinitite.**
1254

1255

1256 Trinitite glasses are a record of the first atomic bomb blast on July 16, 1945. The
1257 detonation of the 21 kilotons plutonium bomb and the resulting fireball melted/vaporized the
1258 arkose/arkosic sand desert and formed a large crater glazed with green fused silica sand. The
1259 explosion released an extreme high amount of energy, with an estimated yield of 9-18 kt, and
1260 average temperature of 8430 K (Hermes & Strickfaden 2005). The heating rate was extremely
1261 high with a very short duration of ~ 3 s and this explain the glass formation and why some
1262 minerals (zircon and quartz grains) were only partially melted (Hermes & Strickfaden 2005).

1263 The green fused silica sand was first described by Ross (1948) that, based on the optical
1264 properties, reports the occurrence of two different melts. In general, he described trinitite as a
1265 highly vesiculated pale bottle green glass that present distinct flow lines. One of the glass phases
1266 has a refractive index (RI) between 1.51 and 1.54 and derives from feldspars, clay fraction and
1267 accessory minerals (calcite, hornblende, augite). The second amorphous phase has a lower
1268 index of refraction (close to 1.46), indicating that this material is nearly pure SiO₂ glass derived
1269 from the fusion of the quartz component of the arkosic sand. Ross (1948) observed also the
1270 occurrence of small glass areas with glasses red or grey in color, both containing copper, and
1271 suggested the it was a contamination from the explosion device.

1272 Since the nuclear explosion, most of the studies have been done on the radioactive
1273 nuclides present in the materials (*e.g.* Atkatz & Bragg 1995; Eby *et al.* 2010; Parekh *et al.* 2006;
1274 Wallace *et al.* 2013). Indeed, the migration of actinides at historical test sites are considered
1275 very important, since it is closely related to the waste management and to the storage of high-
1276 level nuclear waste (Hu *et al.* 2008; Pacold *et al.* 2016; Tompson *et al.* 2002, 2006). However,
1277 for long time the study from Ross was the only mineralogical investigation available on trinitite
1278 (except a magnetic susceptibility study by Glass *et al.* 1987; see below). Only in the last decade
1279 the interest for these materials raised again, mainly because of the nuclear forensic applications,

1280 and many studies on the mineralogy, petrology, geochemistry and on the average Fe or Ti redox
1281 state have been published. For example, [Eby et al. \(2010\)](#) report the occurrence of four different
1282 type of glass:

1283 1) glassy and vesicular green top surface coated with sand grains (the top part of the trinitite
1284 layer) ([Figure 16](#));

1285 2) a Cu-rich glass (red trinitite) containing metallic chondrules;

1286 3) green trinitite (scoriaceous fragments);

1287 4) trinitite ejecta, which include aerodynamically shaped droplets, beads, and dumbbell glasses.

1288 The latter were discovered a few years after the detonation as mm-sized specimens.

1289 Supposedly, part of the molten material was transported by the hot gas cloud and spread over a
1290 wide area into beads and dumbbell shapes ([Eby et al. 2010](#)). The red trinitite contains several

1291 metallic chondrules having Cu, Fe and Pb deriving from the explosion (blast tower and trinity
1292 device), confirming the hypothesis of [Ross \(1948\)](#). The most common material is the green

1293 trinitite that consists of vesiculated and amorphous scoria-like fragments, and it is extremely

1294 heterogeneous. Light microscopy, SEM and BSE analysis show the presence of euhedral heated

1295 sand grains, large flow bands and strong chemical variability ([Bellucci et al. 2014](#); [Eby et al.](#)

1296 [2010, 2015](#); [Fahey et al. 2010](#)) (see [Figure 17](#)). The more recent analysis confirmed the early

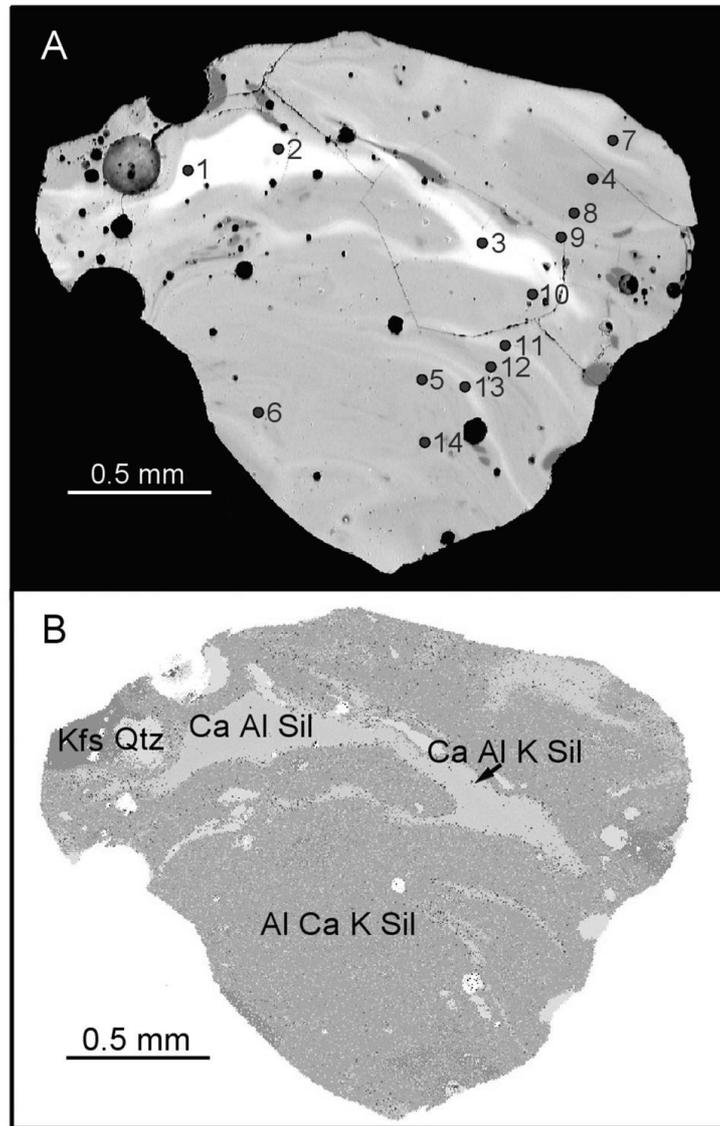
1297 observation done by [Ross \(1948\)](#) on the bulk chemistry. Trinitite glasses have two major glass

1298 compositions, one essentially consisting of lechatelierite, and a second one, depleted in silica,

1299 with a much higher variability (SiO_2 range = 55 - 80 wt.%). Moreover, [Eby et al. \(2015\)](#) reports

1300 the occurrence of planar deformation features in quartz grains, a typical evidence of shock

1301 metamorphism in impactites and fulgurites.



1302

1303 **Figure 17** (a) Backscattered electrons (BSE) and (b) QEMSCAN (Quantitative Evaluation of Minerals by
 1304 SCANNing electron microscopy) images for a green trinitite fragment. Besides the partially melted K-
 1305 feldspar (Kfs) and quartz grains (Qtz) there are evident flow structures, and the occurrence of different
 1306 glass compositions. The brightest areas are enriched in calcium content. Numbers represents the point
 1307 analysis. Figure from Eby *et al.* 2015 (American Mineralogist. 2015;100(2-3):427-441. doi:10.2138/am-
 1308 2015-4921) MSA copyright.
 1309

1310 Because of the resemblance between trinitite ejecta and distal impact ejecta, trinitite
 1311 glasses have been compared to tektite and microtektites (Glass *et al.* 1987). The similarities
 1312 between the two products were confirmed, especially regarding the water contents (between
 1313 0.01 and 0.05 wt.%) and the predominant reducing conditions ($Fe^{3+}/Fe_{tot} < 0.1 \pm 0.1$; Glass *et*
 1314 *al.* 1987; Giuli *et al.* 2010b; Eby *et al.* 2015). The study of the mineralogy and chemical
 1315 variability of the glass samples has constrained the conditions occurring during the atomic

1316 detonation (Pressure-Temperature (P-T) regime), with temperatures on the order of ~ 1870 K
1317 and pressure ≥ 8 GPa (Eby *et al.* 2015). A recent study highlights also the occurrence of nano-
1318 structures in zircon grains, and in particular the presence of baddeleyite fibers surrounding the
1319 core of unaltered zircons (Lussier *et al.* 2017). These fibrous that seem to irradiate from the
1320 zircon grains, are not in physical contact with the core. Based on Raman, TEM and electron
1321 diffraction data, the P-T regime was constrained to $T > 1770$ K and $P < 10$ GPa (Lussier *et al.*
1322 2017). Because of the disagreement between temperatures estimated by trinitite glasses and
1323 fireball (~ 1800 K vs. ~ 8000 K, respectively), it was suggested that the very short duration of
1324 the nuclear event has a major impact on the physical properties registered by glasses and
1325 minerals (Eby *et al.* 2015).

1326 The study of the glasses formed during nuclear events provides information on the type
1327 of device that was detonated and its origin, thus it has nuclear forensic applications. Recently,
1328 Molgaard *et al.* 2015 produced synthetic nuclear glasses, comparable with trinitite glasses, as
1329 surrogates that could be used to simulate a variety of scenarios and could be used as a tool for
1330 developing and validating (nuclear) forensic analysis methods.

1331

1332

1333 **5. Glass properties**

1334
1335 The study of the glasses described in the previous sections can provide important
1336 information of the formation mechanisms of these amorphous materials, and on the related
1337 events. In particular, the information provided by the study of shock metamorphism materials
1338 have implications well beyond earth sciences, since they involve also planetary science,
1339 biogeochemistry, and materials science.

1340 By looking at different scales, it is possible to obtain information on the glass average
1341 redox conditions during fast cooling, on the short and medium range silicate network, on liquid
1342 immiscibility phenomena, and on the macroscopic properties (optical, physical, magnetic and
1343 so on...). All these data can be used to constrain the regimes of temperature and pressure during
1344 formations, evaporation and condensation stages, the timescale of the processes, and the
1345 thermal history of glasses. Altogether with the data from experimental studies (*e.g.* shock
1346 experiments) they are the base for simulations and numerical models of crater formation. In
1347 the following we report some of the characteristic of the amorphous materials and a compilation
1348 of Fe redox state and Raman spectroscopy data, collected on a variety of non-magmatic glasses.

1349

1350 **Liquid immiscibility**

1351 A common feature of many natural glasses is the occurrence of two coexisting different
1352 melt phases, with features similar to liquid-liquid immiscibility. For example, in the previous
1353 sections, the presence of compositional heterogeneities on a variety of scales has been described
1354 for several impact related glasses (*e.g.* Libyan desert glass, Darwin glass), fulgurites and
1355 trinitite. Moreover, the occurrence of ferromagnesian droplets coalescing into a felsic
1356 amorphous matrix has been reported also for pyrometamorphic materials (*e.g.* [Capitanio et al.](#)
1357 [2004](#)).

1358 Multicomponent silicate systems are stable over wide ranges of P-T-X- fO_2 (pressure,
1359 temperature, bulk composition, oxygen fugacity), nevertheless, they might exhibit
1360 immiscibility at particular conditions (*e.g.* [Hamann et al. 2018](#); [Hudon & Baker 2002](#); [Roedder](#)
1361 [1992](#)). Silicate-silicate liquid immiscibility is a common phenomenon observed in many natural
1362 igneous melts on Earth, and in lunar mare basalts (as a late-stage immiscibility), including
1363 meteorites (*e.g.* [Hudon et al. 2004](#); [Roedder 1978](#)) and is a fundamental research field in glass
1364 and ceramic industry (phase separation; see [Vogel 1994](#) for a review). The silicate-silicate (or
1365 carbonate) fluid immiscibility arises from differences in physical properties between two
1366 separate phases, and usually the immiscibility is characterized of coexisting felsic and mafic
1367 components; however, the chemical distribution is highly dependent on the duration of melting,
1368 the cooling rate and ambient atmosphere (reducing or oxidizing conditions). In highly energetic
1369 events, there are dynamic pressure and temperature conditions that allow the physical and
1370 chemical interaction (respectively, mingling and mixing) of very different target materials;
1371 however, because of the timescale of these processes, equilibrium conditions are hardly
1372 achieved, opposite to magmatic processes where a complete homogenization can be attained
1373 (for more information on Magma mixing see chapter [Morgavi, this volume](#)).

1374 Liquid-liquid immiscibility arises from the different structural role of the cations in the
1375 glass. For instance, the main cation building the network in natural compositions is Si^{4+}
1376 (altogether with AlO_4 tetrahedra), which is bonded by four oxygen ions to form a tetrahedral
1377 coordination; each oxygen is shared by neighboring tetrahedra (bridging oxygens) to form a
1378 three-dimensional network held together by strong covalent bonds. The other cations present in
1379 a natural composition have different structural roles, and may either aid or disrupt the silicate
1380 network, thus they are called, respectively, network-formers or network modifiers (see [Chapter](#)
1381 [XX](#), this volume). According to [Veksler \(2004\)](#) the cations that the most influence the liquid-
1382 liquid immiscibility fields are aluminum and alkali/alkali earths, and in turn, the peraluminous

1383 or peralkaline nature of the melts, and cations that present high field strength tend to concentrate
1384 in the Si-poor phase.

1385 Liquid immiscibility seems to be a very common feature of glasses deriving from highly
1386 energetic events, and recently, [Hamann *et al.* \(2018\)](#) discuss the importance of liquid
1387 immiscibility in impact processes, and the mechanisms that lead to the formation of melt
1388 heterogeneities and unmixing of silicate phases. In particular, besides the classical liquid-liquid
1389 immiscibility (rising from phase separation of a melt), the authors define other two main
1390 mechanisms that could produce the heterogeneities observed in silica impact glasses: i)
1391 mingling or incomplete mixing due to the short time-scale associated with highly energetic
1392 events; ii) emulsification of incompatible melts (deriving from the target rocks and impactor),
1393 without mixing. Moreover, [Hamann *et al.* \(2018\)](#) report that, despite the huge differences from
1394 phase separated volcanic rocks (both in terms of composition and T-P-time conditions), impact
1395 glasses show a very similar major element partition between phases.

1396 Heterogeneity in glasses from high energetic events have been described in detail for
1397 Libyan Desert glass, blue zhamanshinites, and also trinitite and fulgurite glasses. In all
1398 specimens the occurrence of amorphous nanospherules, as a result of silicate–silicate liquid
1399 immiscibility, have been observed ([Feng *et al.* 2019](#); [Pratesi *et al.* 2002](#); [Zolensky & Koeberl
1400 1991](#)). Other immiscibility like features can be found also in suevite glasses from Ries crater
1401 where mixture of carbonate impact melt and silicate melt have been reported ([Graup 1999](#)).

1402

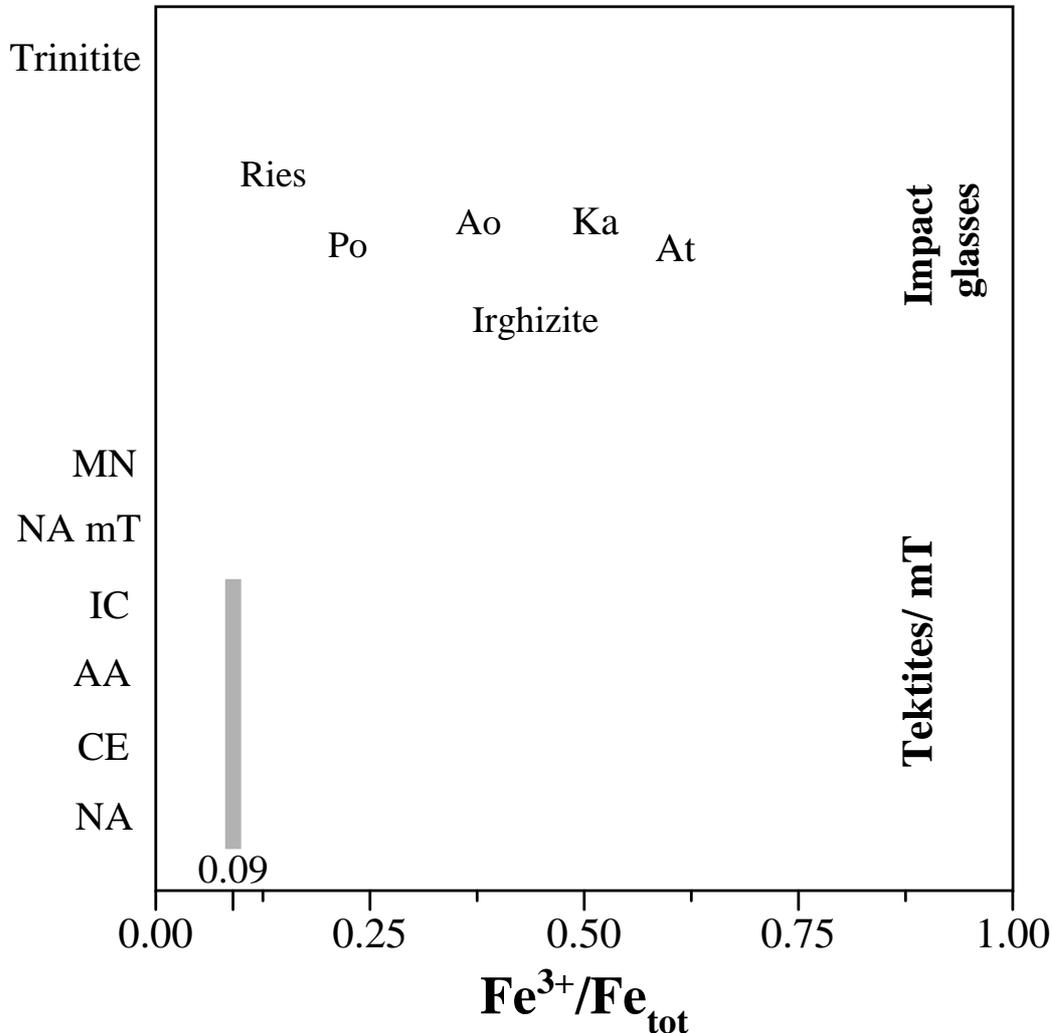
1403

1404 **Reduced iron species**

1405 As discussed in the previous sections, the highly reducing conditions of glasses/rocks
1406 formed from highly energetic events have many repercussions, not only on the formation
1407 mechanisms of the samples themselves, but also on the understanding of the processes that
1408 occurred during the heavy meteorite bombardment, and on the evolution of the early Earth or
1409 to understand planetary impact cratering processes and differentiation. Among the
1410 characteristics common to all distal ejecta there is the occurrence of very high amount of
1411 reduced iron species Fe^{2+} in the melts, and several mechanisms have been proposed to explain
1412 the presence of reduced species, and its preservation during the “travel”, and the quench (*e.g.*
1413 see [Engelhardt *et al.* 1987](#); [Ganino *et al.* 2018](#); [Lukanin & Kadik 2007](#)).

1414 Iron is by far the most common multivalent element in all natural amorphous materials,
1415 with two valence states (Fe^{3+} , Fe^{2+}) that are stable over a large range of P-T conditions. In the
1416 study of glasses derived by impact or other highly energetic events, the occurrence of reduced
1417 iron has become one of the common markers to differentiate those samples from volcanic rocks.
1418 Moreover, in order to constrain some of the condition of formation of natural glasses, the study
1419 of iron valence has been one of the most common tools. An overview of the average Fe valence
1420 in these samples is provided in [Figure 18](#), where $\text{Fe}^{3+}/(\text{Fe}^{2+} + \text{Fe}^{3+})$ (hereafter, $\text{Fe}^{3+}/\text{Fe}_{\text{tot}}$) ratios,
1421 are reported for many natural glasses. The data for tektite and microtektites where obtained by
1422 several techniques (wet chemistry, Mossbauer, XAS; see below for details). Tektites, from the
1423 different strewn-fields, aside some extreme values reported in the literature, all show an average
1424 Fe redox ratio $\text{Fe}^{3+}/\text{Fe}_{\text{tot}} \sim 0.09$ (± 0.10 ; vertical grey line), thus indicating a similar thermal
1425 history and similar P-T- $f\text{O}_2$ conditions, despite their different geographical distribution. In
1426 Muong-Nong-type tektites (MN) and North American microtektites (NA mT), the large range
1427 of $\text{Fe}^{3+}/\text{Fe}_{\text{tot}}$ ratios is, in the first case, typical of the layered tektites, and in the second one, an
1428 unique case among microtektites (see [section 3.1](#)). Distal ejecta have considerably lower

1429 Fe^{3+}/Fe_{tot} ratios than impact glasses, or Fe valences associated to average crustal target rocks
 1430 (the possible precursor material).
 1431



1432
 1433 **Figure 18:** Ranges of Fe redox state (as Fe^{3+}/Fe_{tot}) of tectites and microtektites from the different strewn
 1434 fields are compared to the Fe^{3+}/Fe_{tot} of impact glasses (proximal from Zhamanshin, Popigai, Kara,
 1435 Auouelloul, Atacama and Ries craters) and trinitite glasses. For tectites, both the extreme values and the
 1436 average one (~0.09) are reported. North American microtektites and Muong-Nong tectites are the only
 1437 distal ejecta specimens that show a broader range of Fe redox states (see section 4), with values that overlap
 1438 the proximal impact glasses.
 1439

1440 Several techniques have been used to infer the Fe oxidation state. The earlier works
 1441 reported quite broad ranges regarding the Fe^{3+}/Fe_{tot} with values between 0 and ~ 0.5. The
 1442 analyses were based on magnetic susceptibility or, more commonly, on wet-chemistry
 1443 methods (e.g. Bouška 1994; Bouška & Povondra 1964; Cuttitta *et al.* 1972; Schnetzler & Pinson

1444 1964; Schreiber *et al.* 1984; Thorpe *et al.* 1963). Early spectroscopic data (Mössbauer) were
1445 obtained for a large dataset (tektites from various locations by Evans & Leung 1979; Fudali *et*
1446 *al.* 1987) finding no detectable ferric iron in one case, and up to 0.12% in the second case.
1447 Lately, very similar results on the average Fe redox state were obtained (*e.g.* Dunlap 1997;
1448 Rossano *et al.* 1999). Mössbauer spectroscopy, respect wet chemistry, provided additional
1449 information on the Fe local environment. The early studies reported a coordination environment
1450 for ferrous iron around 6, with a minor presence of 4-fold coordinated species, but more recently
1451 it was reported the main presence of ferrous sites with coordination between 4 and 5 (trigonal
1452 bipyramids) (*e.g.* Dunlap *et al.* 1998; Dunlap & Sibley 2004; Rossano *et al.* 1999; Volovetsky
1453 *et al.* 2008).

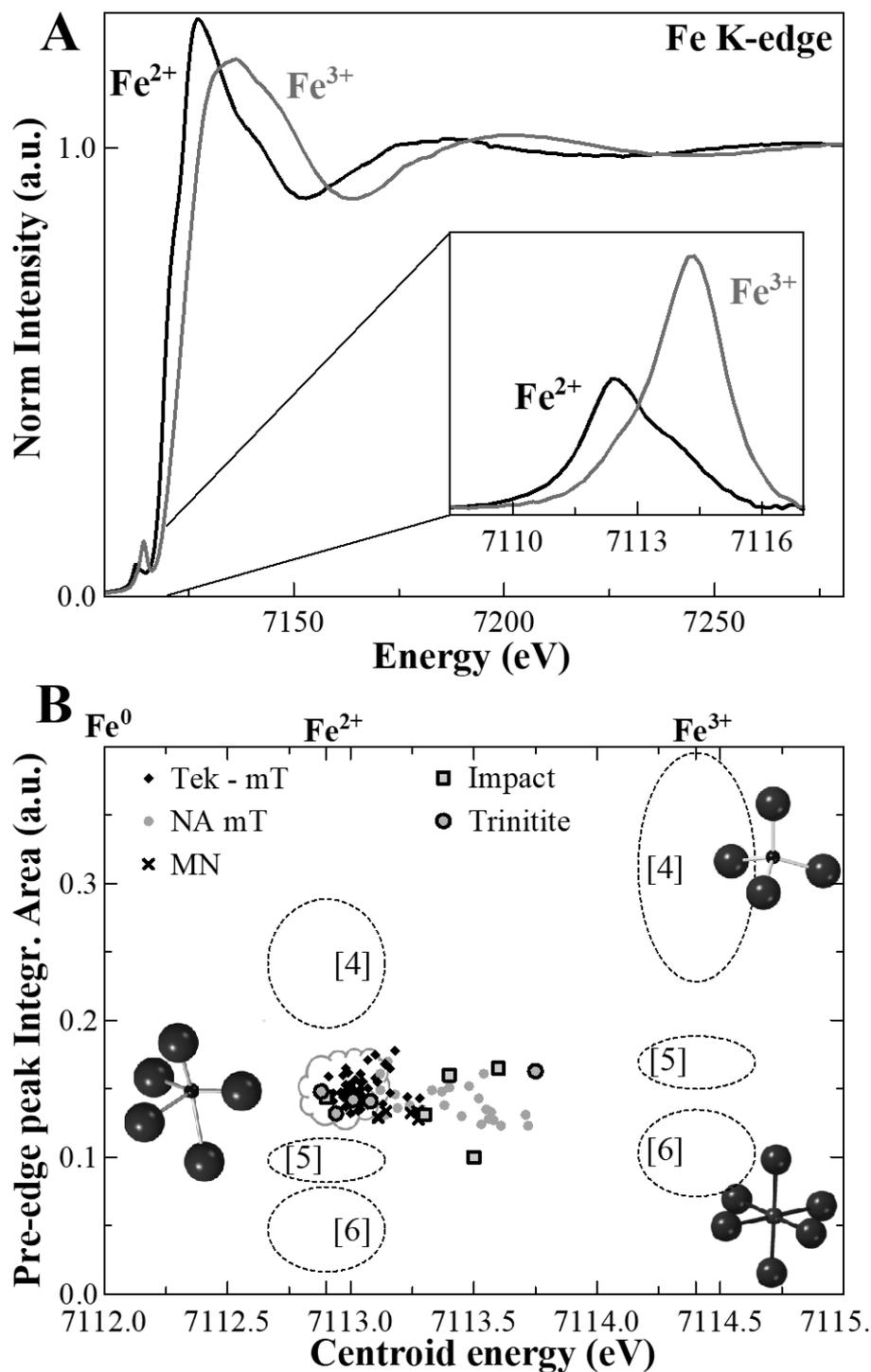
1454 Another spectroscopy technique that is element selective is X-Ray Absorption
1455 Spectroscopy (XAS). The study of the absorption edge region (XANES), including the small
1456 feature before the whitenline called pre-edge peak, and the extended energy region (EXAFS)
1457 provide information on an element average valence state and coordination number. This
1458 technique has been used to study Fe, Al, Ti structural environment in many natural amorphous
1459 materials, to provide insights on pressure and oxygen fugacity conditions during cooling.
1460 Because iron is the most common multivalent transition elements in natural melts, it has been
1461 extensively studied both in natural and synthetic systems, thus, accurate information can be
1462 derived by the study of the Fe K-edge XAS spectra of tektites, impact glasses and so on. XAS
1463 spectra related to reduced Fe²⁺ and oxidized Fe³⁺ glasses show differences in their energy
1464 position, as well as in their shape, with the spectrum related to oxidized species shifted toward
1465 higher energies (Figure 19A). The small feature before the absorption edge (inset in Figure 19A)
1466 is the pre-edge peak that presents different energy positions and intensities depending,
1467 respectively, on Fe oxidation state and average coordination number (local symmetry), and the
1468 deconvolution of this small feature for many crystalline compounds allowed to create the

1469 variogram reported in **Figure 19B** and to derive quantitative information on the Fe structural
1470 role; indeed, the intensity of the pre-edge is related to the number of first neighbors and site
1471 geometry, whereas the centroid position is related to the average valence. For a review on the
1472 technique and on studies related to iron in silicate glasses, melts and minerals, the readers are
1473 referred to [Calas & Petiau 1983](#); [Giuli *et al.* 2002, 2012](#); [Henderson *et al.* 2014](#); [Mottana 2004](#);
1474 [Wilke *et al.* 2001, 2004, 2007](#).

1475 The results obtained from the study of the Fe K-edge pre-edge peak for an extensive
1476 dataset of tektites and microtektites provide very consistent results, with all distal ejecta having
1477 similar values both as average oxidation state and coordination environment. The pre-edge peak
1478 variogram in **Figure 19B** shows that tektite and microtektites cluster in a position that is
1479 compatible with the presence of highly amount of reduced species ($\geq 90\%$ Fe²⁺), and an average
1480 coordination between 4 and 5. Trinitite glass as well has a similar behavior and the iron
1481 structural environment resemble that of distal ejecta. Impact glasses from different localities
1482 have much higher centroid energy values, corresponding to higher amount of trivalent iron (data
1483 in **Figure 19B** from [Giuli *et al.* 2002, 2010a, 2010b, 2013b, 2014a, 2014b](#)). Beside the analysis
1484 of the Fe pre-edge peak, the analysis of the extended region of the Fe K-edge XAS spectra
1485 allowed to derive average $\langle\text{Fe-O}\rangle$ distances of 2.00 ± 0.02 Å, values compatible with the
1486 presence of ferrous species with coordination between 4 and 5 ([Giuli *et al.* 2002](#)).

1487

1488



1489

1490 **Figure 19:** A) XAS at the Fe K-edge for two glasses having reduced and oxidized iron species. The inset
 1491 shows in detail the background subtracted pre-edge peaks (data from [Cicconi et al. 2015](#)). B) Centroid
 1492 energy position against the Fe pre-edge peak integrated intensity. The dashed ovals represent different
 1493 coordination environments of the iron species in minerals. This variogram is obtained from the study of the
 1494 Fe K-edge pre-edge peak of several tektite, microtektites from the different strewn-fields, impact glasses
 1495 and trinitite glasses. Nearly all the distal ejecta cluster in a small area, here highlighted by the cloud shape.
 1496 Data from [Giuli et al. 2002, 2010a, 2010b, 2013b, 2014a, 2014b](#). Figure modified after [Cicconi et al. 2020](#)

1497

1498

1499

1500 **Glass structure**

1501 Most of the natural materials discussed in this chapter are SiO₂-rich aluminosilicate
 1502 glasses. The study of the silicate network arrangement can provide information on glass
 1503 polymerization, temperature and pressure regimes, as observed in **Figure 3**. One of the most
 1504 studied phases is SiO₂, either the amorphous phase or the crystalline polymorphs, and for sake
 1505 of clarity, we compare the properties of amorphous SiO₂ with those of the other crystalline
 1506 phases in **Table 4**. There are 11 known SiO₂ polymorphs formed depending on the different
 1507 pressure and temperature systems, and here we report the most common ones in natural glasses.
 1508 Quartz, tridymite and cristobalite are low pressure polymorphs, whereas coesite and stishovite
 1509 are the high pressure ones (see **Figure 3**). The arrangement of the silica is always the same in
 1510 all polymorphs, namely having a silicon surrounded by four oxygens arranged in a tetrahedra
 1511 (4-fold coordinated), except stishovite where silicon is surrounded by six oxygens.

1512

1513 **Table 4: Some properties of lechatelierite glass and crystalline SiO₂ polymorphs.**

name	Density (g/cm ³)	Refractive Index	<Si-O> (Å)
Lechatelierite	2.20	1.458	1.62
α-quartz	2.65	1.549	1.61
β-quartz	2.53	1.537	1.59
α-tridymite	2.25	1.471	1.59
α-cristobalite	2.32	1.484	1.60
Coesite	2.92	1.596	1.61
Stishovite*	4.29	1.81	1.78

1514 * silicon coordination is 6.

1515

1516 Raman spectroscopy is a very powerful technique that allow to derive information on
 1517 the silicate melt short- and medium-range orders, thermal history, pressure regimes, and to
 1518 provide an overview of sample heterogeneity. Moreover, Raman has been widely used on
 1519 obsidian specimens in order to get information of the provenience (determination of the

1520 geographic origin) of archeological obsidians (*e.g.* [Bellot-Gurlet et al. 2004](#)). Besides magmatic
1521 glasses, some of the first studies that exploit Raman technique to understand the origin of other
1522 natural glasses were done as early as in the 80s, and by using polarized Raman spectra the
1523 authors tried to derive information on the thermal history of Libyan Desert Glasses ([Galeener
1524 et al. 1984](#)), or proximal and distal ejecta ([Jakes et al. 1992](#)). Despite these early investigations,
1525 it is surprising that in the last decades, not so many studies have used this technique to
1526 understand possible modifications in the glass connectivity, but rather to determine the
1527 crystalline phases present, and the peak shock pressure (for instance, see case study in [section
1528 3.2](#)).

1529 In the following, we will report a series of Raman spectra collected for several glasses
1530 described within this chapter. A short description of the main vibration modes of silicate glasses
1531 is reported for sake of clarity; however, for a comprehensive description of Raman spectroscopy
1532 and on the silicate network short- and medium-range order, the readers should refer to dedicated
1533 text ([Neuvillle et al. 2014](#); [Henderson_Stebbins this volume](#)). Raman spectra for other natural
1534 glasses, including magmatic ones, are reported in [Cicconi & Neuvillle \(2019\)](#).

1535

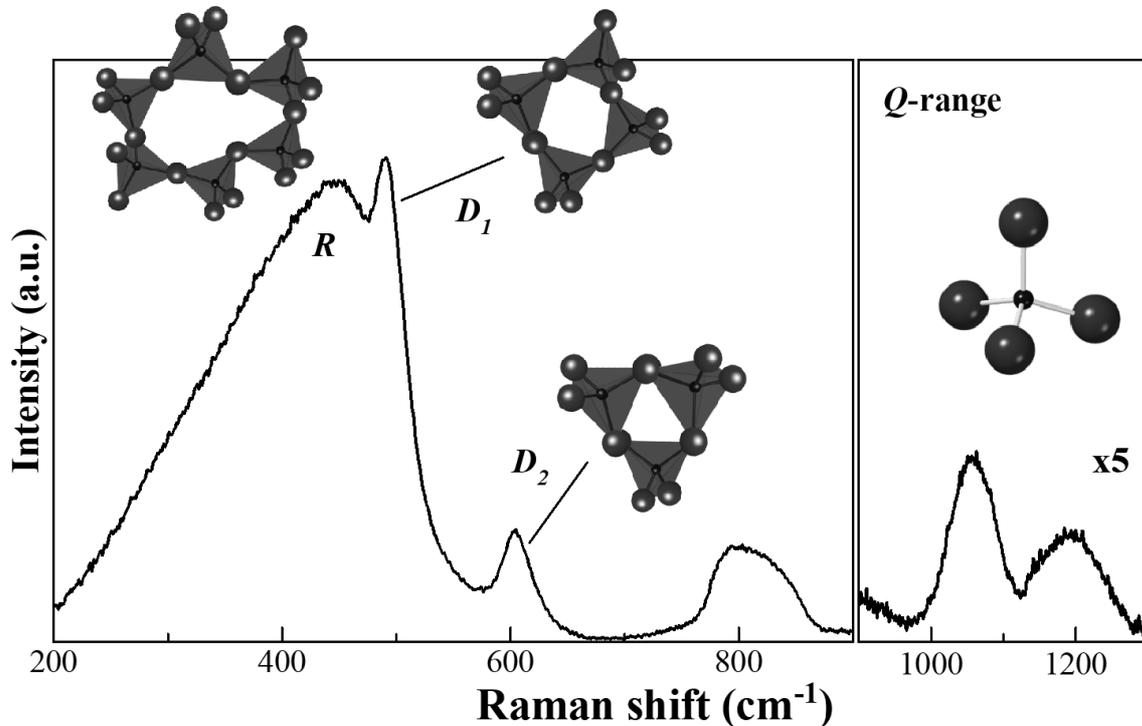
1536 Raman spectra of silicate glasses have the main vibration modes in the 10-1300 cm^{-1}
1537 frequency range, which can be divided in different major regions: a very low wavenumber
1538 region ($< 250 \text{ cm}^{-1}$), which provides information on tetrahedral arrangement (see [Neuvillle et
1539 al. 2014](#) for more details). The frequency range between ~ 400 and $\sim 650 \text{ cm}^{-1}$ can be related to
1540 vibrations of the tetrahedra-rings ([Figure 20](#)), with the main asymmetric band (R) usually
1541 assigned to symmetric vibrations of T-O-T in ≥ 5 -membered rings (*e.g.* T is the Si^{4+} - or Al^{3+} -
1542 centered tetrahedron). Depending on glass polymerization, two additional sharper bands can be
1543 observed ([Figure 20](#)), the so -called defect bands, D_1 ($\sim 490 \text{ cm}^{-1}$) and D_2 ($\sim 600 \text{ cm}^{-1}$), assigned
1544 to breathing modes of 4- and 3-membered rings, respectively. The frequency position and the

1545 shape of the R band strongly depend on the silica network, and for instance, it shifts toward
1546 higher frequencies by densification of SiO₂ glasses (Deschamps *et al.* 2013), or by changing
1547 glass connectivity (*e.g.* by adding network modifier cations) (Neuville *et al.* 2014), thus by
1548 varying T-O-T inter-tetrahedral angles, and T-O average distances. The high wavenumber
1549 region, extending from ~ 900 to 1300 cm⁻¹, contains T-O asymmetric stretching vibrations;
1550 usually these stretching modes are labelled according to T polymerization: Q^{0-4} , where Q
1551 represents the Si(Al) centered tetrahedron, and $0-4$ represents the number of bridging oxygens
1552 (BO). This region is also called Q -range or Q -envelope (Figure 20). By decreasing glass
1553 polymerization (*e.g.* by adding network modifier cations), the Q -range strongly increases in
1554 intensity and shift toward lower frequencies because other bands, related to different Q species
1555 (Q^3 , Q^2) appear in the 900-1200 cm⁻¹ region (*e.g.* Neuville *et al.* 2014).

1556 There are several studies dedicated to the modification occurring to the structure of pure
1557 silica glass upon compression by hydrostatic pressure (*i.e.*, DAC – diamond anvil cell) or shock
1558 experiments. These investigations report that there is a permanent increase in silica glass
1559 density (densification), and for example, the density of a SiO₂ glass increases steadily up to
1560 values 20-21% higher, approaching the density of crystalline SiO₂ (Bridgman & Šimon 1953;
1561 Cohen & Roy 1961, 1965; Susman *et al.* 1991), and for applied pressure below 28 GPa, this
1562 densification is not due to changes of silicon coordination, but rather to structural changes,
1563 mainly related to variations in intertetrahedral angles distribution and the ring statistic, without
1564 variations in Si-O coordination (Deschamps *et al.* 2013; Sugiura *et al.* 1997). Okuno *et al.*
1565 (1999) carried out shock wave experiments on a SiO₂ glass by impacting glass with a steel flyer
1566 (speed up to ~2 km/s) for pressures in the range of 17.8 GPa to 43.4 GPa, and they observed
1567 the maximum density increment (maximum changes in silica network) at 26.3 GPa, The study
1568 of the Raman signals of compressed or shocked glasses has allowed to get many information

1569 on the variations that may occur in the silicate network after high grade shock metamorphisms,
 1570 and to identify the peak pressure values.

1571

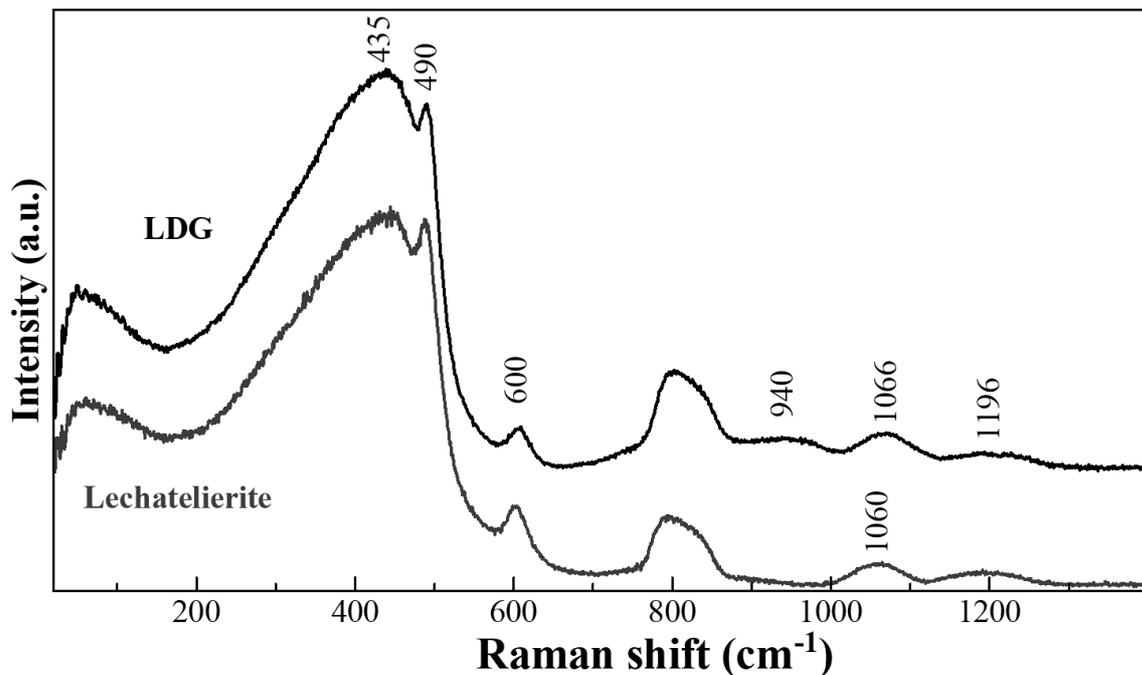


1572

1573 **Figure 20:** Background subtracted Raman spectrum of a commercial pure SiO₂ glass. The
 1574 different bands can be attributed to specific Si-O-Si and Si-O vibration modes. See text for
 1575 details on band labels.
 1576

1577 A common feature of many natural glasses is the occurrence of lechatelierite, that is
 1578 amorphous SiO₂, or glass phases enriched in silica content. For example, different Libyan
 1579 Desert Glass (LDG) clear fragments have been analyzed by using a 488 nm laser excitation,
 1580 and all Raman signals present vibrations with frequency positions and intensities very similar
 1581 to that of a lechatelierite glass, including the two sharp peaks related to the breathing modes of
 1582 4- and 3-membered rings (defect lines). Some authors used the relative intensity of the defect
 1583 bands in the Raman spectra to get information on the thermal history of natural glasses
 1584 (Champagnon *et al.* 1997; Galeener *et al.* 1984), and for example Galeener *et al.* reported a
 1585 fictive temperature (Tf) of 1000 ± 50 °C for LDG, and suggested that the molten material cooled
 1586 down to temperature below Tf in a period ranging between a few minutes and days.

1587 The whole region in the frequency range $700 - 1300 \text{ cm}^{-1}$ of the LDG Raman spectrum
 1588 has a higher background, and all fragments analyzed present a contribution around 940 cm^{-1}
 1589 that is not present neither in the commercial SiO_2 glass, or in the lechatelierite spectrum (**Figure**
 1590 **21**). Because this band is not related to luminescence (electronic transitions), it must be related
 1591 to other cations present in the Libyan Desert Glasses. Stretching modes related to 4-fold
 1592 coordinated Al^{3+} will mainly influence the relative intensity and frequency position of the band
 1593 at $\sim 1200 \text{ cm}^{-1}$, and the intensity of the defect lines because of the changes in the ring statistic;
 1594 hence, the higher background and the band at $\sim 940 \text{ cm}^{-1}$ might be related to other (relatively)
 1595 abundant cations, such as Ti^{4+} and Fe^{3+} species ($\sim 0.1 \text{ wt.}\%$).

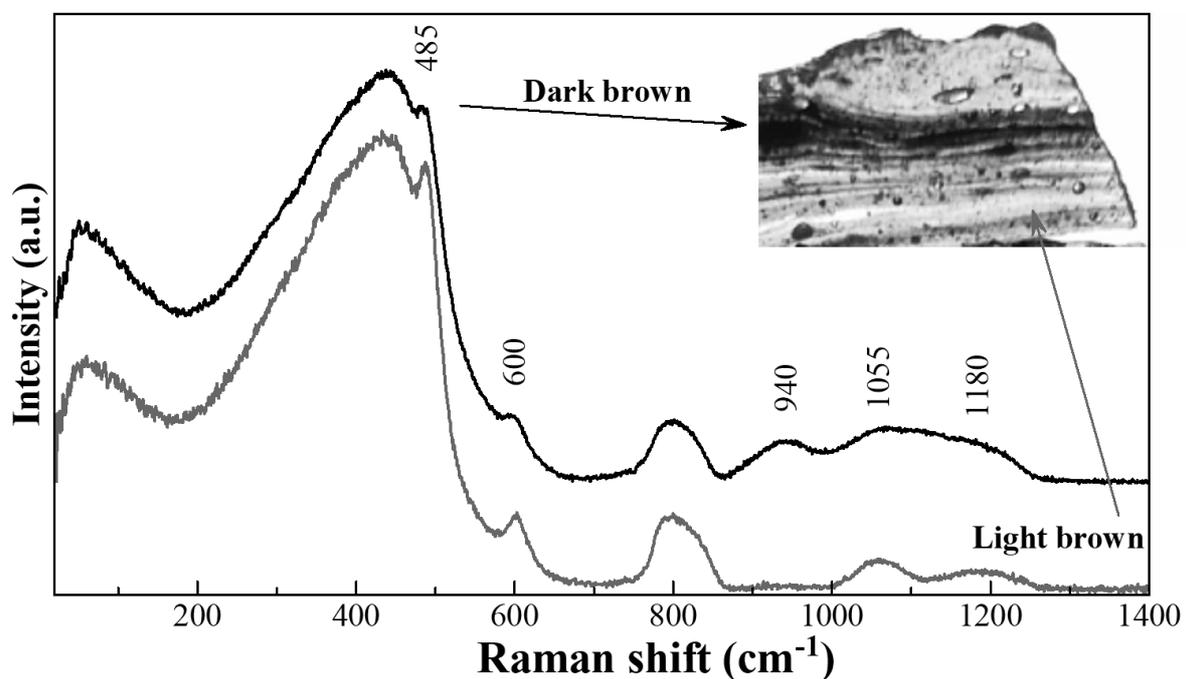


1596

1597 **Figure 21: Raman spectra in the frequency range $20-1400 \text{ cm}^{-1}$ of lechatelierite and Libyan**
 1598 **Desert Glass (LDG). The frequency position of the main vibrations is reported.**
 1599

1600 Darwin glasses are chemically varied, and the Raman signals reflect the heterogeneity
 1601 of these samples. Indeed, depending on the portion analyzed it is possible to get vibrations
 1602 almost completely related to amorphous SiO_2 (*i.e.*, lechatelierite) or vibration modes that
 1603 indicate a more depolymerized silicate network (**Figure 22**). For instance, the layered structure

1604 of the Darwin glass shown in **Figures 7 and 8** have vibrations related to a highly polymerized
 1605 glass in the light layering, whereas the darker layers have lower intensities of the defect lines
 1606 D_1 and D_2 , and stronger bands in the high frequency region, most probably related to higher
 1607 amounts of non-bridging oxygens (NBO). The frequency region between 860 and 1400 cm^{-1} in
 1608 the dark portion has an integrated area almost three times higher than in the light layers, and
 1609 these modifications can be attributed to change in the bulk chemistry (see **Table 2**). In the dark
 1610 glass portion there is also the appearance of an additional contribution around 940 cm^{-1} .

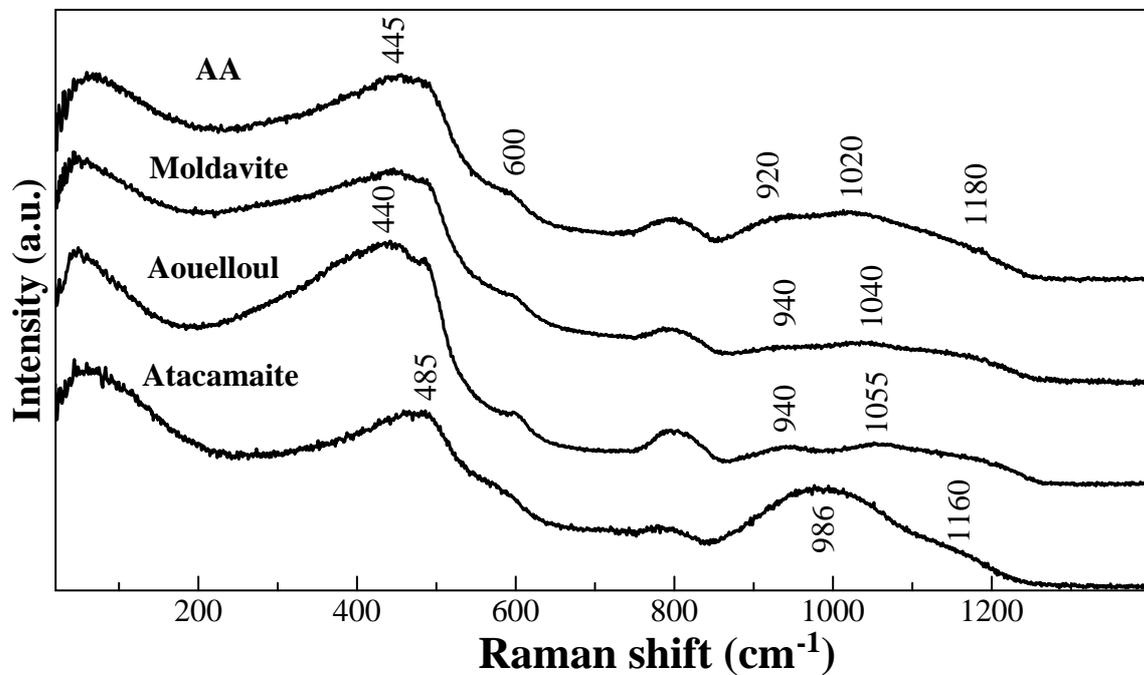


1611

1612 **Figure 22:** Raman spectra in the frequency range 20-1400 cm^{-1} of the light and of the dark
 1613 brown layers in a Darwin glass specimen. The frequency position of the main vibrations is
 1614 reported.
 1615

1616 Distal ejecta and impact glasses have high SiO_2 contents (typically > 65 wt.%; see
 1617 **Tables 1 and 2**), thus the typical vibrations of silica rich glasses are clearly observed, even if,
 1618 sometimes, the defect lines are barely visible (**Figure 23**). Again, the high frequency regions
 1619 show further contributions, and are very different from the lechatelierite glass, or more in
 1620 general from the SiO_2 -richer LDG samples. Atacamaite sample is the most different one, with
 1621 strong variations in the frequency position of the vibrational bands, and an intense Q -envelope.

1622 These modifications on the Raman spectrum of atacamaite glass reflect the average bulk
1623 chemistry of the glass, and especially the relatively low SiO₂ content, and the high Al₂O₃, FeO
1624 and CaO contents.

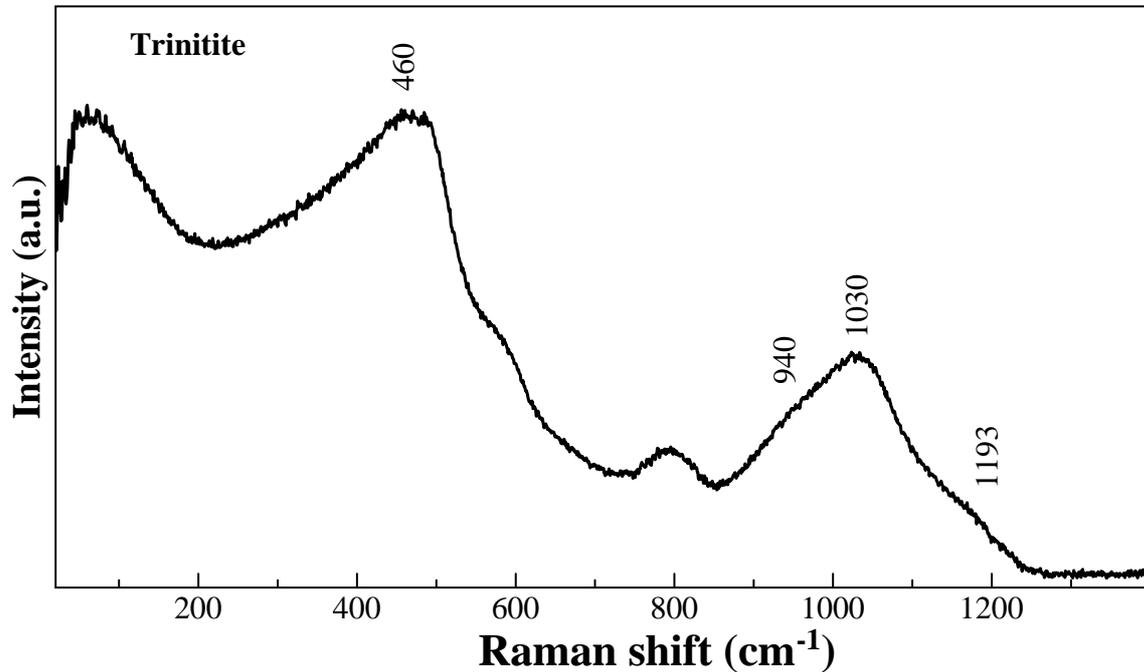


1625

1626 **Figure 23:** Raman spectra in the frequency range 20-1400 cm⁻¹ of several impact related glasses:
1627 two tektites, respectively from the Australasian (AA) and Central European (moldavite) strewn
1628 fields, and two proximal impact glasses (Atacamaite and Aouelloul). The frequency position of
1629 the main vibrations is reported.
1630

1631 **Figure 24** report the Raman spectrum of a bottle green trinitite specimen, and only the
1632 most homogeneous inner glass portion was considered. At the microscale, trinitite specimens
1633 are chemically highly heterogeneous (see [section 3.3](#)) with SiO₂ contents that range between ~
1634 90 and 60 wt.% ([Eby et al. 2010](#)). In the specimen available the Raman spectra collected do not
1635 show Si-rich portions, and the majority of the data collected points to an aluminosilicate glass
1636 composition with SiO₂ contents << 75 wt.%. The high frequency range has very intense and
1637 broad bands, with the highest contribution peaking at ~1030 cm⁻¹. Depending on the glass
1638 portion considered the intensity of the shoulder at ~940 slightly varies, and the R band decreases

1639 its intensity. These small variations can be related to the different proportions of network
1640 modifier cations, and in turn, to the SiO₂ content variability.



1641
1642 **Figure 24:** Raman spectra in the frequency range 20-1400 cm⁻¹ of a trinitite sample. The
1643 frequency position of the main vibrations is reported.
1644

1645
1646 The Raman spectra shown here are just an overview of the variability of the glass
1647 structures, and demonstrate the potentiality of using Raman spectroscopy, especially for
1648 investigations at the microscales. Despite the chemical complexity of a multicomponent natural
1649 glass, the vibrational bands can be interpreted based on the study of simplified synthetic
1650 compositions, and it is possible to correlate the degree of polymerization to the Raman features.
1651 All changes occurring in the low frequency region can be mainly associated to the changes in
1652 the bulk composition, and more in detail, on the Si content that drives the glass network
1653 polymerization. For an almost constant bulk composition, it would be possible to use the glass-
1654 memory effect and study the variations of the intensity of the defect lines, and the variations in
1655 the position of the main R band, in order to relate them to the pressure or the thermal history of

1656 the glasses. The high frequency region contains the T-O stretching modes related to Si^{4+} , but
1657 also of other network former cations, such as Al^{3+} . The variations observed in all glasses,
1658 compared to the bands of lechatelierite, could be related to the different proportions of network
1659 modifiers that creates higher amounts of NBO, and charge compensator cations, that will
1660 influence the covalency character of the bonds. Moreover, other cations, such as the 4-fold
1661 coordinated Fe^{3+} , can also provide stretching vibrations in the high frequency region. For
1662 instance, in aluminosilicate and silicate glasses, both natural and synthetic, it has been reported
1663 that the bands between 800 and 1200 cm^{-1} are very sensitive to the average iron valence state
1664 (e.g. [Di Muro et al. 2009](#); [Di Genova et al. 2016](#); [Le Losq et al. 2019](#)). Thus, in the case of
1665 Darwin brown glass, and atacamaite, the contribution observed could be related to the presence
1666 of higher amounts of 4-fold coordinated Fe^{3+} .

1667

1668 We have described several natural glasses not deriving from volcanic processes, and we
1669 observed that glasses formed as a consequence of highly energetic event share many
1670 characteristics, such as:

- 1671 • Compositional similarities (e.g. the presence of lechatelierite)
- 1672 • Liquid-liquid immiscibility
- 1673 • Highly reduced species
- 1674 • Shocked minerals

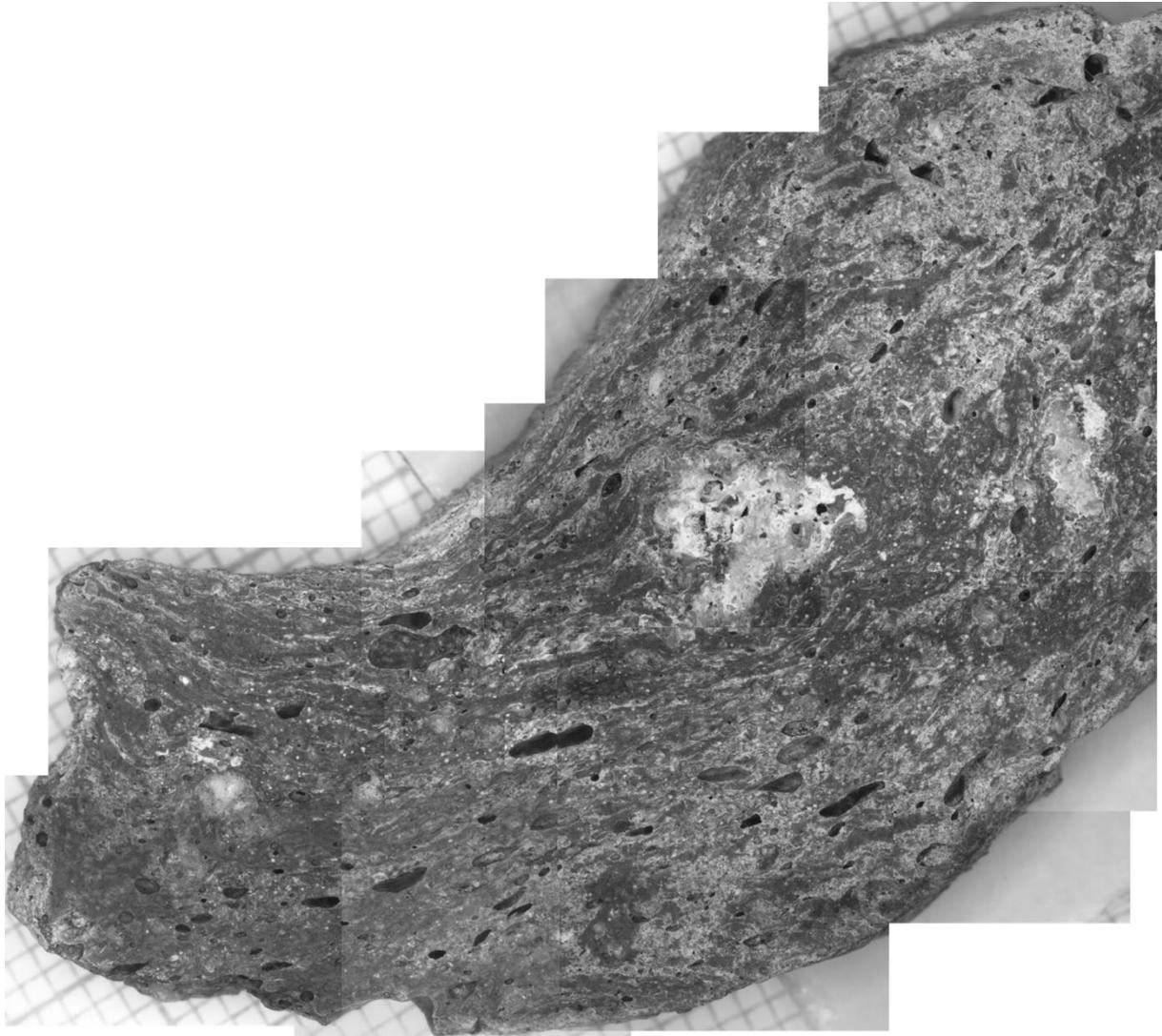
1675 The study of fulgurites and trinitite glasses may serve as a proxy for impact-induced
1676 mineralogic and petrologic changes, and to better understand the formation of glasses from
1677 highly disequilibrium conditions.

1678

1679 **NOT INCLUDED – no time for mapping.**

1680

1681



1682

1683 **A polished slice of suevite (glass bomb) shows the flow lines and the elongated vesicles, along with inclusion**
1684 **of heterogeneous fragments, and glassy pockets (darker spots).**

1685

1686 Flow lines; glass pocket; elongated micro and macro bubbles;

1687 Polished interior section of suevite “glass bomb” showing large (mostly elongate) vesicles that follow the flow lines.

1688

1689

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