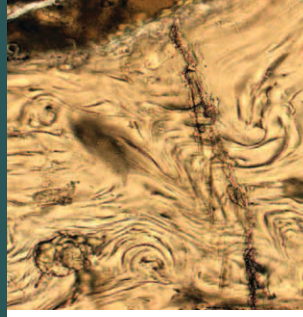
Shock Metamorphism of Minerals



Impact melt glass with a schlieren-type flow texture in a suevite sample from the Chesapeake Bay impact crater, USA

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The hypervelocity impact of extraterrestrial objects causes unequivocal changes in the target due to extreme deformation rates, pressures up to hundreds of gigapascals, and postshock temperatures that may even vaporize silicates. This article introduces the basic principles of shock compression, as required to understand the formation and geological significance of shock-metamorphic effects in minerals. Special emphasis is placed on the formation of high-pressure phases such as stishovite and diamond as well as on the decomposition of carbonates.

KEYWORDS: shock effects, high-pressure phases, degassing of carbonates, Hugoniot curve, planar deformation features, shock metamorphism

INTRODUCTION

Shock metamorphism is the progressive breakdown of the structural order of rocks and their constituent minerals. It is caused by a short pulse of high dynamic pressure, which occurs exclusively in the contexts of natural impacts, nuclear and chemical explosions, and shock and cratering experiments. In shock metamorphism, pressure, temperature, and strain and quench rates commonly exceed by orders of magnitude conditions prevailing during endogenic processes. For example, strain rates during shock compression range from 10^6 s^{-1} to 10^9 s^{-1}, and are thus at least 5 orders of magnitude greater than those attained in even the strongest earthquakes. However, these exceptional physical conditions prevail in nature for just a few seconds —even during the impact of kilometer-sized projectiles— resulting in the formation of diagnostic shock-metamorphic effects. We concentrate here on instructive examples to explain the response of minerals to shock compression; this response is distinctly different from the transformation and deformation of minerals in all other geological processes. For a comprehensive description of effects related to impact cratering, the reader is referred to French and Koeberl (2010).

MINERALS IN A TRANSIENT STATE OF "SHOCK"

During an impact event, minerals adapt abruptly to the extreme conditions prevailing in a shock wave (FIG. 1). Rock-forming minerals such as quartz and feldspar are compressed to almost half of their volumes at pressures of ~100 GPa within nanoseconds to seconds. The shock front propagating with supersonic velocity through a crystal and separating compressed from uncompressed material may only be a few lattice planes wide. As a result, equilibrium states are usually not attained under shock compression, and minerals respond by the formation of specific disequilibrium features, which are collectively known as "shock-metamorphic effects."

This fact can be illustrated on the basis of the phase diagram shown in FIGURE 2. In shock compression, quartz instantaneously reaches pressure–temperature states on the Hugoniot curve that are largely within the stability field of the high-pressure phase stishovite. Yet instead of forming stishovite, quartz reacts by producing mechanical twins, planar fractures (PFs), planar deformation features (PDFs), and diaplectic quartz glass (= densified silica glass).

Most shock-metamorphic effects were first recognized in the 1960s using optical microscopy, spectroscopic techniques, and X-ray diffraction of experimentally and naturally shocked minerals. The physical nature of some of the effects, however, was not understood, and some effects were not even known until transmission electron microscopy (TEM) was employed for the characterization of shocked minerals (e.g. Goltrant et al. 1992; Leroux et al. 1994; Stöffler and Langenhorst 1994). Based on these data and their interpretation, shock effects and processes occurring during shock compression and decompression can be divided into the following classes:

Deformation: formation of dislocations, planar microstructures (PFs and PDFs), mechanical twins, kink bands, and mosaicism (a phenomenon reflecting the breakup of a crystal into small domains)

Phase transformations to high-pressure phases and diaplectic glass

Decomposition into two or more phases (e.g. a solid residue and a gaseous phase)

Melting and **vaporization** of the entire mineral (which is subsequently either quenched or condensed as glass or polycrystalline aggregates)

Deformation and transformation features are caused by the transient shock compression. Decomposition, melting, and vaporization, however, are temperature-dominated effects that develop during unloading to ambient pressure. This is because loading and release follow different paths

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Box 1 WHAT IS A SHOCK WAVE?

The propagation and properties of shock waves are best illustrated by the train model shown in Figure 1. The collision of a moving train (projectile) with a static train (target) leads to deformation and compression of both the moving and the static train—but in opposite directions. The two boundaries between compressed and uncompressed wagons are the shock fronts, which propagate with the shock velocity, U_s. The compression of wagons leads to a displacement of the initial collision front, that is, the boundary between the hitting locomotive and the last target wagon. As a consequence, material flows behind the shock wave into the target, at a particle velocity, u_p, that is distinctly smaller than the shock velocity, U_s. As the process proceeds, more and more wagons are engulfed by the compression until the shock wave in the projectile is reflected as a rarefaction wave at the free rear side of the last wagon in the projectile. This leads to a backward acceleration of rear wagons corresponding to the ejection of material in impact cratering. As the rarefaction wave propagates in already compressed material, it moves faster than the shock wave in the target. Consequently, the shock wave will be overtaken at some stage by the rarefaction wave. This process is the decompression of material that proceeds from the surface of the projectile into the target, where the shock wave finally decays completely.

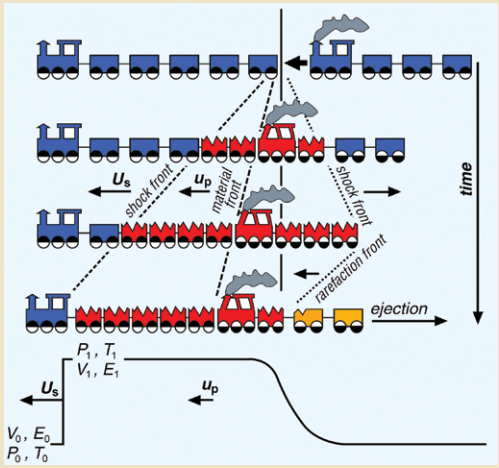


Figure 1 Analogy of the collision of two trains, illustrating the formation and propagation of shock and rarefaction waves (modified after Langenhorst 2002). *P*, *T*, *E*, and *V* are pressure, temperature, internal energy, and volume, respectively, in the uncompressed (state 0) and compressed (state 1) matter.

(Fig. 2), such that part of the pressure–volume work remains as waste heat. No single mineral shows all the listed effects, as the shock behavior of a mineral largely depends on its composition and crystal structure. Quartz, for example, with its three-dimensional covalent bonding between SiO_4 tetrahedra, cannot respond by dislocation glide, whereas dislocations are readily activated within nanoseconds in olivine, which, as a nesosilicate, lacks strong interconnection of SiO_4 tetrahedra (Langenhorst et al. 1999a). Similarly, volatile-bearing minerals respond to shock by dissociation but develop less deformation and transformation phenomena than anhydrous silicates.

DIAGNOSTIC SHOCK INDICATORS

Only one macroscopic diagnostic shock indicator exists: shatter cones. These structures display striations radiating from an apex in fine-grained rocks (see Reimold and Jourdan 2012 this issue). At the microscopic scale, the most commonly observed shock-metamorphic effects are planar

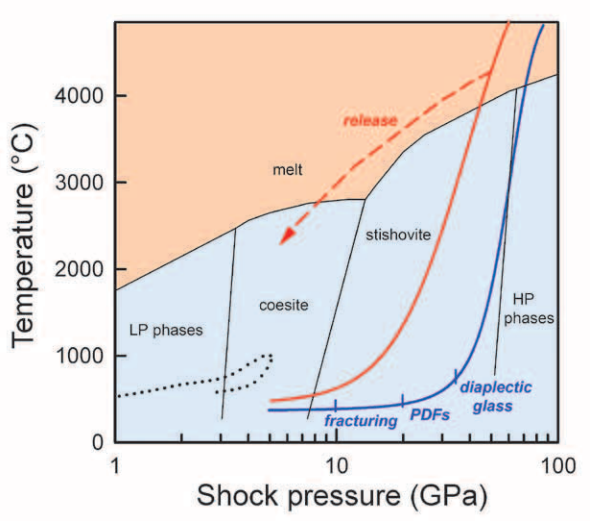


Figure 2 Phase diagram of silica showing the Hugoniot curves of quartz (blue) and sandstone (red). Hugoniot curves are nonisothermal equations of state, named after the French physicist Pierre-Henri Hugoniot, who derived the famous Rankine-Hugoniot equation in 1889. These curves depict all states that a material attains under shock compression. The Hugoniot curve of sandstone demonstrates that porous materials reach higher temperatures in the shock wave than nonporous materials (Wünnemann et al. 2008). The release path is followed during unloading to ambient pressure. The dotted line corresponds to the *P*–*T* path of diamond-bearing eclogite. Low-pressure (LP) phases are low- and high-quartz, tridymite, and cristobalite. High-pressure (HP) phases are $CaCl_2$-type and α-PbO_2-type polymorphs. PDFs = planar deformation features.

structures in rock-forming silicates. Planar microstructures in quartz (Fig. 3) are very valuable for the analysis of shock metamorphism because (1) quartz is a widespread and stable mineral in the terrestrial environment and (2) the planar microstructures, PFs and PDFs, can be easily documented with the optical microscope. PFs are sets of parallel, open fissures with a typical spacing of about 20–30 µm; they are low-pressure (<10–20 GPa) phenomena identical in appearance to cleavage. PFs in quartz are particularly diagnostic of impact because quartz does not possess cleavage planes.

PDFs are probably the most prominent, shock-induced deformation effect. For example, the discovery of PDF-bearing quartz in the Cretaceous–Paleogene (K–Pg) boundary-event bed was the key for the acceptance of this layer as a global impact horizon (Bohor et al. 1987). As revealed by TEM analysis, PDFs are thin (<1 µm), amorphous lamellae with the same composition as the host crystal (Fig. 3a, b, c), developed along rational crystallographic planes (Goltrant et al. 1992; Stöffler and Langenhorst 1994). PDFs occur in many rock-forming silicates, but they are found typically in quartz and feldspars, which develop PDFs at pressures between 10–15 and 35 GPa. Most PDFs in quartz are oriented parallel to rhombohedral planes such as $\{10\bar{1}3\}$ and $\{10\bar{1}2\}$ (for details on the measurement of PDF orientations and the indexing of their planes, see, for example, Langenhorst 2002). The orientation of PDFs in quartz is sensitive to pressure and is regarded as the most robust shock barometer. Even post-shock annealing and alteration merely convert the PDFs into the "decorated" type, preserving the PDFs in their original orientation (Grieve et al. 1996). The decoration is the result of recrystallization of the amorphous lamellae and precipitation of numerous water bubbles. PDFs should

not be confused with decorated subgrain boundaries, which are usually not perfectly planar (Fig. 3D); the latter are the result of tectonic deformation and recovery.

At shock pressures of about 30–35 GPa, quartz and feldspars transform into diaplectic glasses. For these minerals, complete collapse of the crystal lattices to glass in the solid state is the only possible response to rapid shock compression. The speed of the shock process inhibits the time-consuming, reconstructive, solid-state transformations of quartz and feldspars to their high-pressure polymorphs, stishovite (Fig. 2) and lingunite. Shock experiments have shown that the transformation of quartz and feldspars to diaplectic glass occurs gradually over a wide pressure range, starting with the formation of PDFs at 10–15 GPa and terminating with the complete change to glass at 35 GPa. This gradual change also involves a continuous decrease in the refractive indices and densities of shocked quartz as a function of increasing pressure (Langenhorst and Deutsch 1994).

HIGH-PRESSURE PHASE TRANSFORMATIONS IN EXPERIMENTS AND TERRESTRIAL IMPACT ROCKS

Phase transformations of minerals in shock waves can be studied by experimentally determining Hugoniot curves and by examining natural impactites and materials recovered from shock experiments. Hugoniot curves display kinks that are due to plastic yielding at low pressure (the so-called Hugoniot elastic limit, HEL) and phase transformations at higher pressure. Transformations yield denser phases that are less compressible, thus causing a steepening of the *P*–*V* Hugoniot curve. The nature and mechanism of a phase transformation are usually not obvious from Hugoniot measurements and require examination of recovered materials. For example, despite the lack of spectroscopic or electron-microscopic evidence for the production of even traces of stishovite in shock experiments, it is widely assumed that quartz undergoes a solid-state transformation to stishovite under shock compression, which then completely reverts back to the amorphous state (e.g. Tschauner et al. 2006). Such an interpretation is implausible due to the reconstructive nature of the transition to stishovite, which requires atomic diffusion and rearrangement of the crystal structure from tetrahedral to octahedral

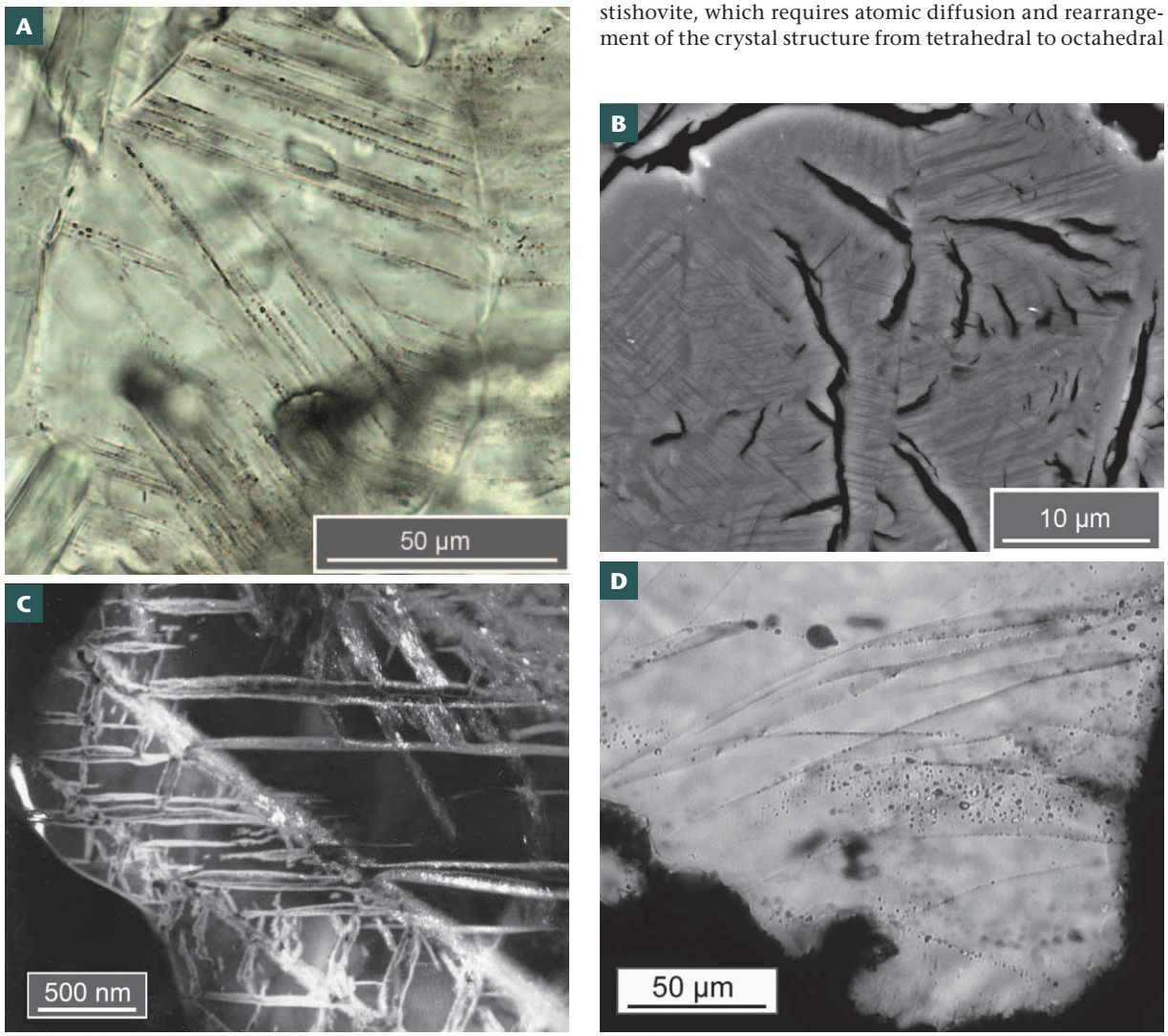


Figure 3 Typical PDFs in quartz: (**A**) An optical micrograph showing two sets of decorated PDFs (drill core sample from the Lake Bosumtwi impact crater). (**B**) A backscattered electron (BSE) image of a quartz grain from a cratering experiment (Ebert et al. 2011) showing, despite the same chemical composition, a contrast between amorphous PDF lamellae (dark grey) and still-crystalline quartz (light grey); black areas are cracks in the sample. PDFs appear darker in BSE images because their density is lower than that of quartz. (**C**) A dark-field TEM image of a shocked PDF-bearing quartz grain from a distal impact ejecta layer in Late Eocene sediments at Massignano, Italy. The layer corresponds to either the Chesapeake Bay or the Popigai impact event. (**D**) An optical micrograph of a quartz grain from the Permian–Triassic boundary at Graphite Peak, Antarctica (Langenhorst et al. 2005). The curved structures are not PDFs; rather they represent subgrain boundaries (curved arrays of dislocations) that formed during tectonic deformation.

coordination of silicon. Such solid-state transitions are simply too sluggish to occur in shock experiments with a pulse duration of <1 µs; even in static high-pressure experiments it usually takes minutes to nucleate stishovite. Instead, the most plausible explanation for the steepening of the Hugoniot curve is the direct transformation to a highly compressed glass.

Traces of coesite and stishovite are, however, widely found in terrestrial impact craters in various settings (French and Koeberl 2010). Both of these high-pressure polymorphs of silica form tiny (<<1 µm) crystals or polycrystalline aggregates in silica glass, either in diaplectic glass or in shock veins or pockets. These regions are hot spots in impactites, where temperatures are high enough to melt quartz under shock compression. Coesite and stishovite are then readily crystallized from the melt, when the release path passes through their stability fields (Fig. 2). A high degree of undercooling is the explanation for the high nucleation rate and the tiny size of the crystals. In strong support of this rapid liquid-crystallization hypothesis is the absence of lattice defects that could be related to solid-state transformation or shock deformation. Instead, coesite shows in TEM studies numerous [010] rotation twins, which are well known as growth defects resulting from rapid crystallization (Leroux et al. 1994). The formation of high-pressure silicates from melt happens at temperatures well above 2000 °C, where the kinetics are much faster than in lower-temperature igneous processes.

The liquidus crystallization of high-pressure silicates (e.g. wadsleyite, ringwoodite, akimotoite, silicate perovskite) is a common phenomenon in shock veins in strongly shocked meteorites (Gillet et al. 2007). In the terrestrial environment, however, postshock annealing and hydrothermal alteration easily obliterate shock veins. It is, thus, not surprising that majorite (a high-pressure polymorph of orthopyroxene) has only recently been discovered in shock veins in a suevite from the Ries impact crater, Germany (Stähle et al. 2011).

Intense research on terrestrial impactites has, however, revealed new high-pressure polymorphic transitions of durable minerals that operate rapidly in the solid state by diffusionless mechanisms. These discoveries include (1) post-rutile transitions to α-PbO_2- and baddeleyite-structured (akaogiite) titania polymorphs (El Goresy et al. 2010), (2) the graphite-to-diamond transformation (Masaitis et al. 1972), (3) the zircon-to-reidite (scheelite-structured) transition (Glass et al. 2002), and (4) the transition of ilmenite to a $LiNbO_3$-structured phase (El Goresy et al. 2010). These transitions are commonly of the martensitic (shear) type, as exemplified by the transition of rutile to the α-PbO_2-structured polymorph (Fig. 4). The rutile structure is described as consisting of approximately hexagonal-close-packed oxygen atoms with titanium atoms occupying half the close-packed rows of octahedral interstices. Shear on oxygen planes in the rutile structure moves the titanium atoms into new positions and results in conversion into the α-PbO_2 structure. Such a martensitic transformation requires a cooperative movement of atoms. However, in reality, the short timescale of shock compression impedes perfect motion, with the consequence that shock-transformed rutile contains numerous planar defects that can be detected only by TEM.

The most spectacular transformation in impact events, one that has attracted the interest of scientists and the public, is the formation of diamonds. Diamonds in terrestrial impact structures were first discovered at Popigai, Siberia (Masaitis et al. 1972), and later in many other impact structures within the territory of the former Soviet Union. An

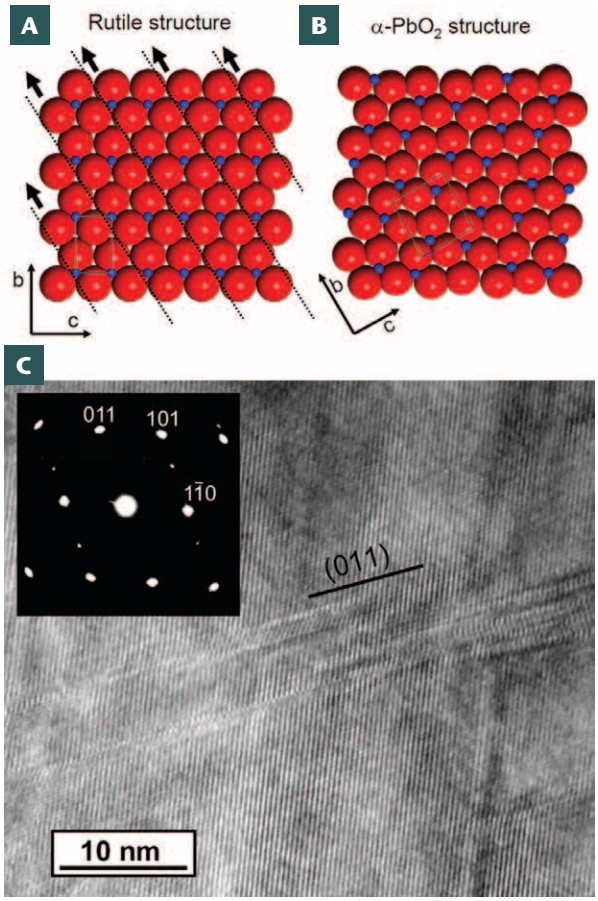


Figure 4 Schematic drawing of the oxygen (red) and titanium (blue) positions in (**A**) rutile and (**B**) the α-PbO_2-structured polymorph. Rutile is transformed into the α-PbO_2 phase by a ½ [01$\bar{1}$] shear on alternate (011) oxygen planes. The high-resolution TEM image combined with the electron diffraction pattern (inset) in (**C**) shows that such a conversion results in planar defects on (011).

early report about the discovery of impact diamonds in suevites from the Ries crater (Rost et al. 1978) was unappreciated until the find was confirmed more recently (Hough et al. 1995). Due to their occasional intergrowth with moissanite (a low-pressure SiC polymorph), the Ries diamonds were first thought to have formed by chemical vapor deposition from the ejecta plume (Hough et al. 1995), but later the occurrence of moissanite was explained by the reaction of diamond with silica-rich impact melt (Langenhorst et al. 1999b). The hexagonal shape of impact diamonds and in situ observations in thin sections substantiate a solid-state transformation of graphite to diamond by a martensitic mechanism (Figs. 5a, b; El Goresy et al. 2001; Langenhorst 2002). This transition involves (1) collective sliding of basal planes of hexagonal graphite (ABAB…) in such a way that a cubic stacking sequence forms (ABCABC…) and (2) compression and puckering of carbon layers along the *c* axis. Due to the martensitic nature of this transition, there is a crystallographic orientation relationship between the crystal structures of the two carbon polymorphs, with $[111]_{dia}$ being parallel to $[0001]_{gr}$. As in the case of the post-rutile transition, the martensitic mechanism yields a very disordered diamond with many planar defects on {111} planes, resulting in unusual physical properties, such as anomalous birefringence (Fig. 5a).

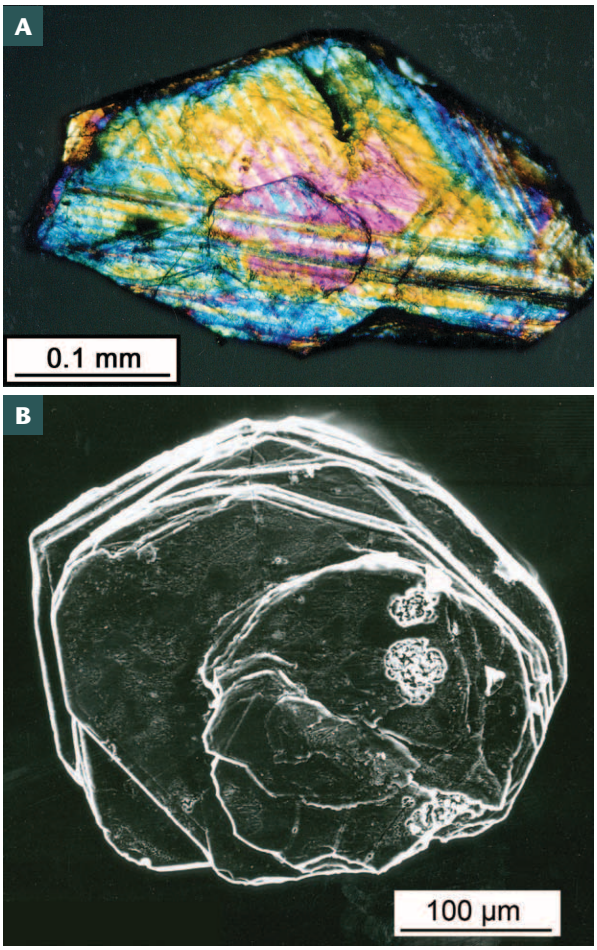


Figure 5 (**A**) Optical micrograph (crossed polarizers) of an impact diamond from the Popigai impact structure, Siberia, showing twin striations, a small hexagonal surface platelet, and unusual interference colors. The birefringence is due to nanoscale defects that internally strain the diamond. (**B**) Secondary electron image of a diamond grain from the Ries crater, Germany. The diamond consists of several overgrown hexagonal platelets. Since this morphology is inherited from graphite, the diamonds can be regarded as pseudomorphs.

SHOCK RESPONSE OF CARBONATES

Sedimentary rocks are common targets in terrestrial impacts. Of the about 180 known impact structures on Earth, about 60 occur in mixed crystalline–sedimentary targets, and about 65 are in sedimentary targets. Thus, carbonates and sulfates play an important role, because they can dissociate during unloading from high dynamic pressure, releasing CO_2 or SO_x as climate-forcing gases. The Chicxulub impact at the K–Pg boundary, which affected a >3 km thick platform sequence of carbonate–sulfate sediments overlying crystalline basement, is the most spectacular example of this process (Pierazzo and Artemieva 2012 this issue).

The stages of progressive shock metamorphism of carbonates at terrestrial impact sites are well documented in a qualitative manner (Osinski et al. 2005; Deutsch and Langenhorst 2007). Shock effects include multiple twinning, PFs, and peak broadening in X-ray diffraction patterns. These effects have been reproduced in shock experiments (Langenhorst et al. 2002), yet may be obliterated easily in nature by annealing and alteration. The transformation of carbonates to densely packed post-aragonite phases was achieved in static high-pressure experiments (e.g. Ono et al. 2007). As these phases cannot be quenched, they are insignificant in the context of natural impact events. In contrast, melting, dissolution of CaO in silicate melts, and complete dissociation of carbonates are extremely important; these are temperature effects, strictly related to the waste heat present in the system during and after pressure release. The presence of high-Ca silica glasses in ejecta from the Chicxulub impact, for example, at the K–Pg boundary in Haiti, provides indirect evidence for decarbonation. Direct evidence for this process in the form of a solid residual phase (periclase, MgO) has so far been reported only by Martinez et al. (1994), from the Haughton impact crater, Arctic Canada. Impact-related melting of carbonates is documented by the coexistence of carbonate and silicic impact-glass spherules in Chicxulub ejecta from Mexican K–Pg sites (Schulte et al. 2010). In addition, carbonate fragments in silicic impact-melt breccias may undergo melting, partly accompanied by dissociation: samples from Chicxulub commonly reveal calcite blebs rimmed by Ca-rich pyroxene, as well as degassing pipes (Deutsch and Langenhorst 2007). The amount of CO_2 released by this obviously slow and long-lasting process is unconstrained.

The phase diagram for calcite ($CaCO_3$) constructed in relation to dynamic compression and following release from shock (Ivanov and Deutsch 2002) demonstrates clearly that the release of CO_2 from shocked carbonates to the atmosphere cannot be deduced from the calcite phase diagram constructed from experiments under thermodynamic equilibrium conditions. During adiabatic unloading from high shock pressure, and hence high shock temperature, calcite passes the *P–T* field of liquid $CaCO_3$ and reaches the field of decomposition. Shock-recovery experiments (Fig. 6) and theoretical considerations indicate that decomposition into a mixture of CO_2 and solid CaO not only requires high residual temperatures but also enough free space to allow expansion of the released CO_2 and diffusion out of the system. If space is strongly restricted, melting will be the major effect on $CaCO_3$, occurring during unloading from high shock pressures.

Two more facts must be taken into account with respect to impact-induced decarbonation in terrestrial impact events: (1) the porosities of limestone and calcareous mud are high enough to lower significantly the critical shock pressure for degassing (cf Fig. 2), and (2) CO_2 can back-react with lime, as shown experimentally by Agrinier et al. (2001).

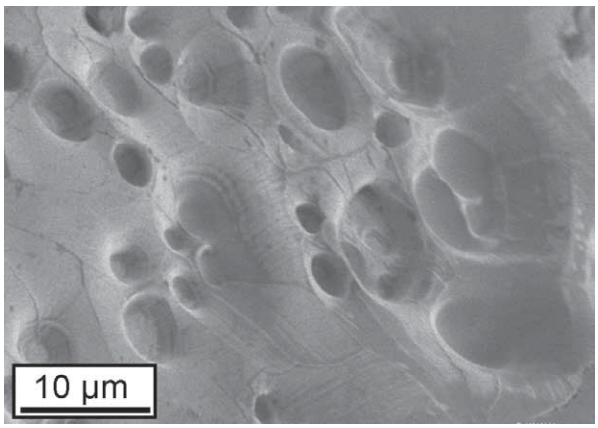


Figure 6 Secondary electron image of a melted single crystal of calcite, produced in a shock-recovery experiment at a nominal pressure of 85 GPa. The vesicles indicate onset of dissociation and release of CO_2, which, however, was suppressed due to confinement by the sample container.

Climate forcing due to impact-released SO_x gas species is an important part of the most plausible scenario for the origin of the K–Pg mass extinction (Pierazzo and Artemieva 2012). Similarly to carbonates, shock experiments with anhydrite ($CaSO_4$) have yielded a wide range of data (Ivanov et al. 2004), but unambiguous evidence for dissociation has so far not been obtained.

CONCLUDING REMARKS

Over the last decade, our knowledge of shock metamorphism, especially transformation processes and the discovery of high-*P* modifications, has been substantially enhanced by high-resolution analytical techniques, among which TEM provides the most important information. Other lines of successful research include dynamic and static loading experiments with sophisticated detection techniques. These experiments provide "pristine" shock effects, allowing the assessment of formation mechanisms and postshock modifications (alteration and annealing) in natural environments. For cases like time-consuming dissociation reactions and phase transformations, experimental limitations exist and modeling may be a more promising approach. Nevertheless, for verifying the impact origin of a geological structure or a sedimentary horizon (Glass and Simonson 2012 this issue), the optical microscope remains the key tool of investigation.

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