Weathering features in shocked quartz from the Ries impact crater, Germany

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Abstract—Shocked quartz from the ejecta of the Ries impact structure has been investigated by analytical transmission electron microscopy (ATEM). Quartz grains display numerous planar fractures (PFs) and planar deformation features (PDFs). Both are partly or fully replaced by a mineral of the kaolinite group (likely halloysite). Its formation involves fluid circulation into the dense fracture networks, dissolution and removal of the amorphous phase initially present in PDFs, and finally, precipitation and crystallization of the kaolinite group mineral from solutions resulting from the chemical alteration of adjacent minerals (feldspars and biotite). Kaolinite group minerals are typical of hydrothermal alteration at low temperature, in humid climate, and under moderately acid conditions and, thus, this alteration may not be directly related to the impact event itself. However, the weathering features were strongly enhanced by the shock-generated microstructure, in particular by fractures that provided pathways for fluid circulation.

INTRODUCTION

Deformations and transformations of rocks that result from shock metamorphism are used as a criterion for the identification of terrestrial impact structures (e.g., Stöffler 1972; Grieve 1991; Grieve et al. 1996). Pressures, temperatures, and strain rates produced in an impact event strongly differ from normal crustal metamorphic and tectonic processes and the resulting shock-induced modifications in rocks and minerals reflect these specific conditions (e.g., Stöffler 1971a; Stöffler 1972; Stöffler and Langenhorst 1994). Because of the widespread occurrence of quartz in the Earth’s crust, shock metamorphism in this mineral has been intensely studied. The main characteristic of quartz shocked in the 10–30 GPa pressure regime is the presence of planar features, which include planar fractures (PFs) and planar deformation features (PDFs). Planar fractures occur in sets of parallel fractures; their formation starts at relatively low shock pressure (below 10 GPa). They are believed to be formed by the extensional forces that accompany the decompression stage after shock wave transition. PDFs consist of multiple sets of parallel, narrow, and closely spaced planar lamellae. They develop at peak pressures above ~10 GPa. Because of their sub-μm size, their exact nature can be determined only by transmission electron microscopy (TEM). They are mostly oriented parallel to low Miller indices planes, such as the basal plane (0001) and the rhombohedral planes \{10\overline{1}1\}, \{10\overline{1}2\}, and \{10\overline{1}3\}. In unaltered shocked rocks or in materials from shock recovery experiments, PDFs in quartz consist of thin amorphous lamellae (e.g., Kieffer et al. 1976; Gratz et al. 1988; Goltrant et al. 1991; Langenhorst 1994). In altered or metamorphosed rocks, the original amorphous material in PDFs is recrystallized. In this case, the original orientation of PDFs is still preserved due to the presence of numerous small fluid inclusions, which have developed along the original planar defect (Goltrant et al. 1991, 1992; Leroux et al. 1994, 1995; Leroux and Doukhan 1996). Fluid inclusions decorating PDFs strongly indicate that recrystallization is water-assisted. Mechanical twinning along (0001) planes also occurs (Goltrant et al. 1971; Leroux et al. 1994), but its identification with the optical microscope is not possible, except where twins are decorated by fluid inclusions.

Despite quartz being a weathering-resistant mineral, post-shock thermal and hydrothermal events in impact craters inexorably modify its original shock-induced microstructure. The observed microstructure is thus the result of the overprint of shock and post-shock histories. Whereas shock microstructure has been intensively studied, weathering of shocked rocks is less well known, although it could provide some important information for the understanding of hydrothermal processes in impact craters.

This paper relates the discovery of new weathering features in shocked quartz from the ejecta layer of the Ries impact structure, as investigated by analytical transmission electron microscopy (ATEM).
SAMPLE LOCALITY AND ANALYTICAL DETAILS

The Ries crater in southern Germany is 25 km in diameter. It was formed about 15 Ma ago (e.g., Stöffler 1974; Stöffler and Ostertag 1983; Engelhardt 1990). Due to its young age, it is one of the best preserved impact craters on Earth. It is also probably the best studied terrestrial impact structure. The investigated quartz grains come from a crystalline breccia sample originating from an ejecta block of moderate shock degree (Engelhardt 1990). The sample was collected in the inner ring, at Appetshofen, east of the town of Nördlingen (sample S289 in Engelhardt and Bertsch 1969; Stöffler 1971b). The parent rock is a biotite plagioclase gneiss containing 19 vol% quartz, 57 vol% feldspar that is nearly completely isotropic (maskelynite), and 24 vol% biotite (Engelhardt and Bertsch 1969). Quartz grains display multiple sets of planar features (Fig. 1), which are typical for shock stage II (Stöffler 1971a, 1971b). Planar features in quartz are frequently filled with crystalline materials of higher refractive index and higher birefringence than the host quartz (Engelhardt and Bertsch 1969). This was originally attributed to the presence of stishovite in PDFs (Engelhardt and Bertsch 1969; Stöffler 1971b).

Quartz grains were first embedded in epoxy and polished sections (30 µm thick) were prepared for examination by optical microscopy. Observations in transmitted light revealed the occurrence of several sets of intersecting planar features (Fig. 1). Three quartz grains apparently showing PDFs filled with high birefringence material were selected and mounted on 3 mm wide copper grids. Electron-transparent TEM thin foils were obtained by Ar ion milling (5 kV, 15°) until specimen perforation. The samples were then lightly coated with carbon. Observations were performed with a Philips CM30 TEM operating at 300 kV. Crystallographic characteristics of the sample were obtained by selected electron area diffraction (SAED), using a double-tilt holder to reach the desired orientation of the foils. The crystallographic orientations of planar defects were determined by orienting the planes parallel to the electron beam. The corresponding lattice planes are then deduced from the corresponding SAED patterns. Chemical compositions were determined by EDS attached to the TEM, in the scanning transmission electron microscope (STEM) mode.

RESULTS

TEM observations show that the quartz grains contain a very high density of planar features (Fig. 2). Several sets of intersecting lamellae are found in all studied grains. Planar deformation features are present at a very high density. They appear in the TEM as straight and parallel lamellae (Fig. 3). Their thickness and crystallographic orientation can be determined only when they are correctly oriented in the TEM, i.e., when the electron beam is parallel to the planar defect. Most PDFs have orientations parallel to the {10\(\bar{1}\)0} and {10\(\bar{1}\)3} planes. They range from 10 to 200 nm in width and are spaced 100–300 nm apart. A number of planar features are spindle-shaped and extend for only 1–2 µm. They are most likely planar fractures. Their orientation is preferentially parallel to the {10\(\bar{1}\)0}, {10\(\bar{1}\)1}, and {0001} planes. Although some PDFs unambiguously contain amorphous material of SiO\(_2\) composition, it is difficult to distinguish PDFs from PFs. Indeed, in some areas both are open planar features, including planar defects along {10\(\bar{1}\)2} and {10\(\bar{1}\)3} planes, giving the impression that the infills have been removed. In most areas, PDFs and PFs are filled with fibrous materials, which frequently have a submicrometer-size, irregular, rod-like morphology. The lengths of these fibers are typically 50–100 nm (Figs. 4a and 4b). Some of them appear only partially filled (Fig. 5). Fibrous microstructure is also found to fill extended areas within quartz grains (Fig. 6). It probably corresponds to weathering of areas that were strongly damaged during the shock event, probably fully...
amorphized. EDS microanalysis shows that the fibrous materials are Al-rich, including those present in planar features. The measured atomic Al:Si ratios range from 0.7 to 1. Minor amounts of K, Ca, and Fe are also detected. The measured composition is close to that of minerals of the kaolinite group, for which the Al:Si ratio is equal to 1. Elemental maps for Si and Al distribution are shown in Fig. 7 (recorded in the STEM mode in an area containing relatively thick, filled planar features). Selected area electron diffraction (SAED) patterns showed intense reflections at 7.2, 4.5, 3.6, and 2.6 Å (Fig. 8). These d-spacings are consistent with lattice spacings of a member of the kaolinite family of minerals, namely $d_{001}$, $d_{020} + d_{(110)}$, and $d_{002}$. The 2.6 Å ring is probably the superposition of several individual reflections, as a large number of d-spacings are consistent with this distance. Based on composition and diffraction patterns only, it is not possible to distinguish a specific member of the kaolinite group. The identification can, however, be made on the basis of distinctive texture. Kaolinite occurs as platelets, whereas halloysite has spherical or tubular morphology (Banfield and Eggleton 1990). As kaolinite family minerals occur in the TEM images as circular cross-sections, halloysite is the preferred choice. Water content in halloysite is highly variable. Depending on the water content, the $d_{001}$ spacing varies from 7 to 10 Å. The 7 Å variety corresponds to dehydrated halloysite (metahalloysite). Water, if present in the starting samples, was probably lost during sample preparation or in the vacuum of the TEM.

**DISCUSSION AND CONCLUSION**

A collision of an extraterrestrial projectile with a planetary surface transfers a high amount of thermal energy to the impact site. Post-shock thermal effects can persist for a long period. Thermal equilibration with neighboring environments can be achieved after, at least, several thousands of years (e.g., Osinski et al. 2001), but this depends on the magnitude of the impact event. The normal geological processes in the area can be strongly affected. The thermal energy deposited in an impact structure can produce hydrothermal activity (e.g., Newsom 1980; Newsom et al. 1986; McCarville and Crossey 1996). During cooling of the structure, a specific hydrothermal circulation system is generated because of temperature difference between the environment and the crater itself. Evidence for impact-induced hydrothermal activity has been documented at the Ries (Newsom et al. 1986, Engelhardt et al. 1995) and Manson, USA impact crater (McCarville and Crossey 1996), impact craters, at Haughton, Canada (Osinski et al. 2001), the Woodleigh impact structure (Uysal et al. 2001), the Kärdda impact crater, Estonia (Kirsimäe et al. 2002), the Lonar Lake impact structure, India (Hagerty and Newsom 2003), and others. The main evidence is the occurrence of clay minerals (such as montmorillonite, illite, Fe-rich saponite, or celadonite), which are believed to originate from hydrothermal alteration at the end of the cooling process, within a temperature interval of 100–200 °C (Newsom et al. 1986; Engelhardt et al. 1995; Uysal et al. 2001; Hagerty and Newsom 2003; Naumov 2002). Another interesting alteration feature is the precipitation of calcite in melt fragments in fallout suevites around the Ries crater (Graup 1999; Osinski 2003). Weathering of minerals and clay mineral formation can also be enhanced by the presence of impact glasses and highly fractured/brecciated rocks, even after the thermal equilibration of the impact site with its environment. It was demonstrated that chemical reactivity of minerals is favored by the presence of shock-induced glasses or internal lattice defects, as well as by new surface areas resulting from intense fracturing (e.g., Boslough 1991). Chemical weathering, in particular aqueous dissolution of shocked materials, is enhanced (Boslough and Cygan 1988; Cygan et al. 1989). For instance, this aspect is nicely illustrated by the preferential
weathering of shocked feldspars to clay minerals along PDFs in samples from the Ries impact crater (French 1998).

Evidence for post-shock thermal effects in shocked quartz has been reported by a number of workers. When unaltered, PDFs in quartz appear as amorphous lamellae (e.g., Gratz et al. 1988; Goltrant et al. 1992; Langenhorst 1994). In weathered impactites, PDFs are found to be strongly modified (e.g., Goltrant et al. 1991; Le roux et al. 1994; Leroux and Doukhan 1996). The most commonly observed effect involves the recrystallization of the amorphous phase and the formation of submicrometer-size fluid inclusions on PDFs (decorated PDFs). Such a microstructure strongly suggests that during weathering some amount of water diffused through the amorphous lamellae and could enhance recrystallization (Leroux and Doukhan 1996). Water is then expelled and forms chains of small fluid inclusions along the original PDF planes. These fluid inclusions become well visible through TEM analysis (e.g., Goltrant et al. 1991; Leroux and Doukhan 1996). When present in high concentration, fluid inclusions generate a brownish coloration of quartz when viewed in thin section (Short and Gold 1996; Whitehead et al. 2002). Both types of information (TEM and optical microscopy) are useful to infer the post-shock thermal history of an impact site.

The weathering features observed in the studied quartz grains strongly differ from textures caused by recrystallization and fluid inclusion formation along PDFs. In the studied quartz grains, minerals of the kaolinite group, likely halloysite, are found within planar features as replacement of PDFs and infill in fractures. The close association of shocked quartz and clay minerals has not been reported so far and brings some new insight about weathering behavior of shocked minerals. Minerals of the kaolinite group are known to result from alteration of feldspars, biotite, or muscovite (e.g., Ahn and Peacor 1987; Robertson and
Weathering features in shocked quartz from the Ries impact crater, Germany

Eggleton 1991; Banfield and Eggleton 1988; Banfield and Eggleton 1990). The kaolinite phyllosilicate minerals are typical of hydrothermal alteration at low temperature, in humid climate, and under moderately acidic conditions. Their formation in the Ries environment was probably not induced by the impact event itself, but was probably favored in highly shock melted/amorphized and fractured rocks. Their formation is likely expressed by the following chemical reaction (an alkali feldspar is given here as precursor mineral):

\[
2\text{Al}_3\text{Si}_8\text{O}_{18} + 2\text{H}^+ + 3\text{H}_2\text{O} \rightarrow 3\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 + \text{K}^+ \tag{1}
\]

During weathering, aluminosilicate minerals are dissolved and hydrolyzed. Cations such as Na\textsuperscript{+}, K\textsuperscript{+}, Ca\textsuperscript{+}, Si\textsuperscript{4+}, and Fe\textsuperscript{3+} are leached away by meteoric water. Minerals of the kaolinite group occur at the late stage of alteration; thereby, halloysite is a transitional phase prior to kaolinite formation. Their occurrence shows that the rock has undergone a high degree of chemical weathering. In the studied shocked rock, formation of kaolinite minerals was probably facilitated by the presence of amorphized feldspars and damaged biotite (remember that the shock stage of the studied rock is II). Their presence within shocked quartz involves fluid circulation, which was rendered possible because of the very dense and interconnected fracture networks in shocked grains, which created permeable pathways. The fluid circulation had two effects. First, it lead to the hydration, dissolution, and removal of amorphous silica that constituted PDFs. Second, these new open spaces and fractures were later sites for precipitation and crystallization of minerals of the kaolinite group. This evidence of weathering in shocked quartz illustrates that shock-induced defects in rocks and minerals strongly favor fluid circulation and alteration. As minerals from the kaolinite group are typical for hydrothermal alteration at low temperature, their formation mechanisms may have no direct relation with the post-shock thermal event itself. It is also possible that this clay mineral formation in shocked rocks is the end member of a series of alteration processes that started at the time of cooling shortly after the impact event.

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