

The effect of water on recrystallization behavior and grain boundary morphology in calcite—observations of natural marble mylonites

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Received 15 March 2004; received in revised form 18 April 2005; accepted 5 May 2005

Available online 19 July 2005

Abstract

Fluids are inferred to play a major role in the deformation and recrystallization of many minerals (e.g. quartz, olivine, halite, feldspar). In this study, we sought to identify the effect of fluids on grain boundary morphology and recrystallization processes in marble mylonites during shear zone evolution. We compared the chemistry, microstructure and mesostructure of calcite marble mylonites from the Schneeberg Complex, Southern Tyrole, Italy, to that from the Naxos Metamorphic Core Complex, Greece. These two areas were selected for comparison because they have similar lithology and resemble each other in chemical composition. In addition, calcite–dolomite geothermometry indicates similar temperatures for shear zone formation: 279 ± 25 °C (Schneeberg Complex) and 271 ± 15 °C (Naxos high-grade core). However, the two settings are different in the nature of the fluids present during the shear zone evolution. In the Schneeberg mylonites, both the alteration of minerals during retrograde metamorphism in the neighboring micaschists and the existence of veins suggest that aqueous fluids were present during mylonitization. The absence of these features in the Naxos samples indicates that aqueous fluids were not as prevalent during deformation. This conclusion is also supported by the stable isotope signature. Observations of broken and planar surfaces using optical and scanning electron microscopes did not indicate major differences between the two mylonites: grain boundaries in both settings display pores with shapes controlled by crystallography, and have pore morphologies that are similar to observations from crack and grain-boundary healing experiments. Grain size reduction was predominantly the result of subgrain rotation recrystallization. However, the coarse grains inside the wet protomylonites (Schneeberg) are characterized by intracrystalline shear zones.

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Keywords: Calcite marble; Mylonitization; Fluids; Microstructure; Recrystallization; Grain boundary morphology

1. Introduction

In many orogenic belts, including, for example, in the Alps (Pfiffner, 1982; Heitzmann, 1987; Burkhard, 1993), Spain (Behrmann, 1983) or Canada (Busch and Van der Pluijm, 1995), marbles often accumulate large amounts of strain in localized shear zones that involve deformation by crystal plastic processes (e.g. Bestmann et al., 2000; Ulrich et al., 2002). Such late-stage shear zones are formed under a variety of thermal regimes and tectonic settings, but often record deformation at relatively low pressures and temperatures (Bestmann et al., 2000). Owing to the extreme

localization of strain, such marble sequences are thought to play a key role in crustal deformation processes, and have often been a subject of field studies (Schmid et al., 1977; Pfiffner, 1982; Behrmann, 1983; Heitzmann, 1987; Burkhard, 1993; Busch and Van der Pluijm, 1995; Badertscher and Burkhard, 2000; Bestmann et al., 2000; Badertscher et al., 2002; Ulrich et al., 2002). The microstructures within these shear zones contain important information on sense of shear, recrystallization mechanisms and final grain size. The stress conditions can be estimated by applying various flow laws, derived from experimental studies (Schmid et al., 1980; Rowe and Rutter, 1990; Walker et al., 1990; Rutter, 1995; Covey-Crump, 1998; de Bresser, 2002; Renner and Evans, 2002; Herwegh et al., 2003). However, despite extensive field and laboratory investigations, many questions remain concerning the mechanical behavior of carbonates, the exact rheology appropriate to describe natural deformation, particularly at very large strains

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(Pieri et al., 2001) and the influence of such variables as pore fluids, second phases and chemical solutes.

One significant consideration for crustal deformation is the presence or absence of water. Experimental deformation of wet aggregates indicate that fluids have a major effect on mechanical properties and microstructural evolution of many minerals, including quartz (Griggs, 1974; Tullis and Yund, 1982; Jaoul et al., 1984; Kronenberg and Tullis, 1984; Hirth and Tullis, 1992; Post and Tullis, 1998), ionic salts (Skrotzki and Welch, 1983; Urai, 1983, 1985; Urai et al., 1986b; Spiers et al., 1990; Spiers and Brzesowsky, 1993; Peach et al., 2001; Watanabe and Peach, 2002; Schenk and Urai, 2004), feldspar (Dimanov et al., 1999), olivine (Mei and Kohlstedt, 2000a,b) or clinopyroxene (Chen and Kohlstedt, 2003; Kohlstedt et al., 2003). In these minerals fluids affect point defect concentrations and diffusion rates, enhance grain boundary mobility or alter dislocation dynamics. At low temperatures, solution transfer processes may operate.

Considering the importance of deformation of marble formations in the processes of mountain building, it is surprising that only a few laboratory studies have been conducted to investigate the influence of fluids on the mechanical properties and recrystallization behavior of calcite.

The results of comparisons between wet and dry samples are somewhat equivocal. Adams and Nicolson (1900) deformed Carrara marble at a temperature of 300 °C in the presence of water, but did not observe differences with similar tests under dry conditions at 300 and 400 °C. Rutter (1974) deformed coarse-grained Carrara marble and fine-grained Solnhofen marble, with and without interstitial water in the range of 20–500 °C. He concluded that the presence of fluids did not significantly affect the mechanical (rheological) behavior of the coarse-grained marble, but that the strength of the fine-grained marble was reduced, at least at low temperatures. The presence of porosity in the Solnhofen marble is significant, and the strength reductions in the Solnhofen may have been related to weakening caused by compaction with an associated increase in pore fluid pressure.

At higher temperatures, in dense, coarse-grained marbles, the presence of water seems to weaken calcite rocks only by a small amount. In a few experiments done on nominally drained samples of synthetic marble at 700 °C, confining pressure of 100 MPa and strain rates ranging from 10^{-3} to $2 \times 10^{-5} \text{ s}^{-1}$, Olgaard (1985) found that wet samples were only a few percent weaker than dry ones. In the most recent study, de Bresser et al. (2005) compared the mechanical properties of pre-dried Carrara marble deformed in axial compression at temperatures between 600 and 1000 °C, confining pressure of 300 MPa and strain rate of 10^{-5} s^{-1} with that of wet, undrained Carrara samples, deformed under the same conditions. At almost all conditions the wet samples were only somewhat weaker

than the dry ones. The final grain shape in the wet samples was somewhat rounder than the dry samples.

Tullis and Yund (1982) and Olgaard (1985) studied the influence of water on the grain growth kinetics in calcite aggregates. The annealing experiments on Solnhofen limestone of Tullis and Yund (1982) at temperatures of 650–1000 °C and confining pressures of 200–1500 MPa indicated faster growth rates of wet compared with the dry samples (but the effect of water is not as pronounced as it is for novaculite (quartz)). In grain growth experiments on fine-grained synthetic calcite, Olgaard and Evans (1988) concluded that normal grain growth in synthetic marble with added water was faster than that in samples containing carbon dioxide inclusions, but slower than that in very pure synthetic marbles with few or no fluid inclusions.

In this study we compare natural marble mylonites that recrystallized under varying pore-fluid conditions. The extent of the interaction between the marble rocks and the pore fluids contained within them has been characterized mainly by stable isotope studies (Burkhard and Kerrich, 1988; Baker and Matthews, 1995; Lewis et al., 1998; Matthews et al., 1999; Kirschner and Kennedy, 2001). We focus on the effect of fluids on the grain boundary morphology and recrystallization behavior. We sampled marble shear zones from the Schneeberg complex (Italian/Austrian Alps) and the high-grade core of Naxos, Greece, both having a similar geological history, but with different amounts of fluids present during late-stage deformation. Across the respective shear zone profiles we looked at the microstructures in thin sections together with chemical data derived from XRF and stable isotopes. In addition, the mylonite samples were broken and investigated under SEM, as done previously for hot-isostatically pressed synthetic marble (Olgaard and FitzGerald, 1993) and natural quartz mylonites from the Simplon Fault Zone (Mancktelow et al., 1998). In a more recent paper, Mancktelow and Pennacchioni (2004) compared natural quartz-feldspar mylonites with variable amounts of water present during mylonitization with respect to the grain boundary structure. They conclude that water-rich fluids enhance the grain boundary mobility in quartz significantly.

2. Geological setting and sampling

2.1. Schneeberg complex

The Schneeberg Complex is a Paleozoic subunit of the Ötztal group (Ötztal–Stubai Complex, Eastern Alps), NNW of Merano (Italy) (Fig. 1), and located between the rest of the Ötztal and the Texel group, which lies to the south (Hoinkes et al., 1987; Schmid and Haas, 1989). From NW towards SE, the Schneeberg Complex is composed of the Monotonous Series, the Heterogeneous Series and the Laaser Series (Sölva et al., 2005). The rocks of the Laaser Series are pure, white marbles and garnet-mica schists

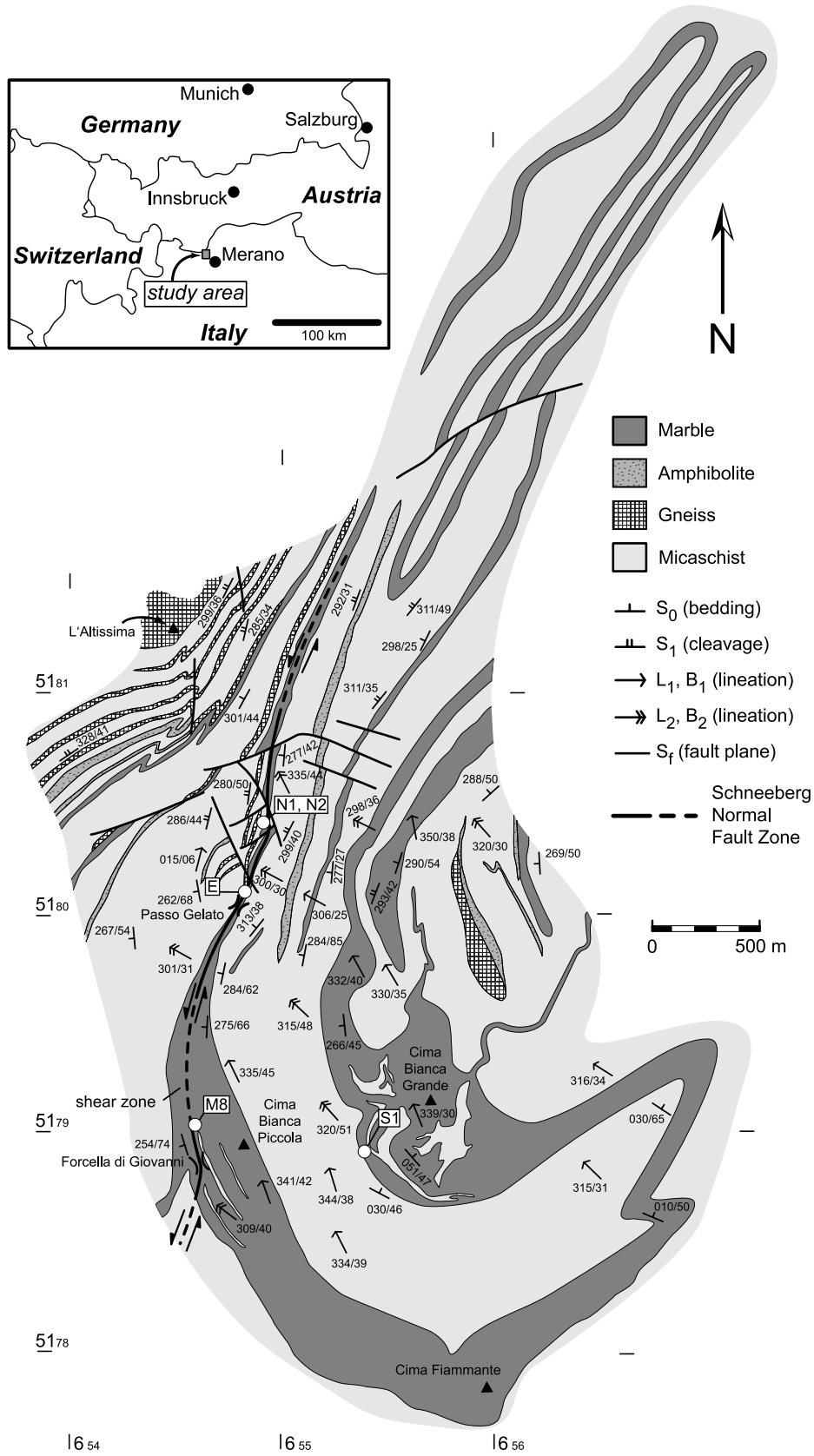


Fig. 1. Geological map of the southwestern tip of the Schneeberg Complex (coordinate system: UTM European Datum 1950). The letters contained in boxes indicate the collection locations for samples listed in Tables 1–5.

intercalated by layers of quartzite, amphibolite and calc-schist.

Whereas the dominant deformation structures of the Ötztal basement are Variscan, the most southeastern part (Schneeberg Complex and Texel group) is overprinted by strong Alpine deformation (Sölva et al., 2001) that formed a well-known set of fold interference patterns with steep fold axes called ‘Schlingen’. Pre-alpine deformation is preserved only as relicts in garnets (Sölva et al., 2001).

The Alpine deformational history is related to the eo-Alpine collision, with the evolution from high-grade to lower greenschist facies metamorphic conditions indicating crustal uplift (Spalla, 1990). Five deformation stages (D_1 – D_5) can be distinguished. Kinematic and geometric shear sense indicators are in agreement with observations of Sölva et al. (2005), who assume that the high pressure rocks of the Texel Complex were continuously exhumed relative to the Ötztal–Stubai Complex, by motion along the NW dipping Schneeberg Normal Fault Zone (SNFZ). This fault zone was active from amphibolite to lower greenschist facies (D_1 – D_4). The microstructure indicates that deformation was accommodated by a mixture of crystal plasticity and cataclastic mechanisms. As temperature decreased, cataclasis became more dominant (particularly in stage D_5) (Sölva et al., 2005), resulting in progressive strain localization in marble units of the Laaser Series.

As a consequence of this localization, the initially coarse-grained marble underwent dramatic grain size reduction.

The area sampled in this study is located in the Schneeberg Complex between the L’Altissima (Hohe Wilde) and Cima Fiammante (Lodner) within the Laaser Series.

We focused on D_4 shear zones, together with their respective marble host rocks and adjacent garnet-mica schists. Sample locations are shown in Fig. 1. See Table 1 for GPS-data and additional observations.

2.2. Naxos high-grade core

Naxos, Greece, the largest Cycladic island in the Aegean Sea (Fig. 2), belongs to the Attic Cycladic Metamorphic Belt (Lister et al., 1984; Feenstra, 1985) and was affected by at least two Alpine regional tectono-metamorphic events (Urai et al., 1990). The first of the two, the main Alpine orogeny (Eocene) was due to the closure of the Mesozoic Pindos Ocean (Hansen and Heide, 1999). This early compressional tectonic phase (D_1) ended 50–40 Ma ago and involved subduction of continental margin material, generation of a nappe pile and regional high-pressure–low-temperature (HPLT) metamorphism (M_1).

That compressional phase was followed by a period of extensional tectonics (D_2) in Early Miocene (Urai et al., 1990). The extensional phase probably accompanied a southward retreat of the N-dipping subduction zone south of Crete (Urai and Feenstra, 2001), during which a back-arc

basin formed with thinned crust, high heat flow, rapid uplift of lower crustal rocks, and intrusion of granitoid magmas (Pe-Piper et al., 1997). Also associated with this extensional phase was a regional greenschist facies metamorphic event (M_{2a} ; ~25 Ma) followed by localized deformation under high-temperature–medium-pressure (HTMP; ~700 °C and 0.6 GPa) metamorphism (M_{2b} ; ~20–16 Ma) (Buick and Holland, 1989). The elongate thermal dome that resulted dominates the structure of the island. The metamorphic zonation is nearly concentric with the dome, as shown in Fig. 2 by six metamorphic isograds mapped in pelitic and bauxitic units (Jansen and Schuiling, 1976). The core of the dome consists of migmatized Hercynian basement, inside of which rafts of coarse-grained, pure, white marble exist. They contain synmetamorphic pegmatites and thin boudinaged amphibolites showing N–S extension during M_2 metamorphism. The core is surrounded by a cover of marble and schist (Hansen and Heide, 1999); together these units are described as metamorphic core complex. This entire core complex is strongly deformed due to a major crustal shear zone that was active during M_2 metamorphism (Urai and Feenstra, 2001). The deformation that resulted is visible as km-scale isoclinal folds with fold axes trending N–S. These folds are coaxially refolded by open, upright folds (Urai et al., 1990). This regional fabric forms the structural dome with its foliation warping over the migmatite core (Urai and Feenstra, 2001).

With decreasing temperatures during further extension and uplift, the deformation was strongly localized in mylonite zones (D_3) inside the marble units and mica schists. These post- M_{2b} mylonites are characterized by extreme grain size reduction (Urai et al., 1990) and are parallel to the local orientation of bedding or to older high-grade schistosity.

The samples of the Naxos core complex for this study were taken inside a marble raft in the high-grade core close to Kinidaros, where very fine-grained post- M_{2b} marble mylonites are embedded in the very coarse-grained calcite marble of high purity (see Fig. 2 and Table 1 for details). As was done for the Schneeberg rocks, oriented samples were taken from the marble mylonites, as well as from the respective host rocks and adjacent mica schists.

3. Observations

3.1. Schneeberg complex

3.1.1. Mesostructures

The Laaser Series rocks consist of marbles, garnet-mica schists and minor intercalated layers of amphibolite, quartzite, dolomite marble and calc-schist. At a distance of ~10 m from the D_4 shear zones, the calcite marble is white, pure, and coarse-grained. The marble is dominantly calcite, but also contains small amounts of quartz and rarely mica. Inside the shear zones, the fine-grained marble

Table 1
Overview of samples described in this study showing the sampling location, sample description and different kinds of chemical analyses used

Samples			GPS ^a		Lithology	Recrystallization	XRF	ICP	Isotopes	Thermometry
			R	H						
Schneeberg complex	S	S-1 ^b	06 55 372	51 79 058	Marble	Protolith				
		S-2 ^b	06 55 372	51 79 058	Micaschist	–				
	N-1	N-1a1	06 54 960	51 80 444	Marble	Protolith			×	
		N-1b1	06 54 960	51 80 444	Marble	Protolith	×		×	
		N-1c1	06 54 960	51 80 444	Marble	Protolith			×	
	N-2	N-2a2a	06 54 942	51 80 486	Marble	Mylonite			×	
		N-2a2b	06 54 942	51 80 486	Marble	Protomylonite			×	
		N-2d	06 54 942	51 80 486	Marble	Mylonite			×	
	E	N-2 h	06 54 942	51 80 486	Marble	Mylonite	×		×	×
		E-1-1	06 54 867	51 80 170	Marble	Mylonite	×	×	×	×
		E-1-2	06 54 867	51 80 170	Marble	Protomylonite	×		×	
		E-1-3	06 54 867	51 80 170	Marble	Protomylonite			×	
		E-1-4	06 54 867	51 80 170	Marble	Protolith		×		
		E-2	06 54 867	51 80 170	Micaschist	–				
		E-3 ^c	06 54 867	51 80 170	Marble	Mylonite			×	×
	M-8	E-4	06 54 867	51 80 170	Marble	Mylonite	×	×	×	
		M-8-1a	06 54 567	51 78 999	Marble	Mylonite	×		×	
		M-8-1b	06 54 567	51 78 999	Marble	Protomylonite			×	
		M-8-2a	06 54 567	51 78 999	Marble	Mylonite	×		×	
		M-8-2b	06 54 567	51 78 999	Marble	Protomylonite			×	
M-8-3a		06 54 567	51 78 999	Marble	Protomylonite	×		×		
M-8-3b		06 54 567	51 78 999	Marble	Mylonite			×		
M-8-3c		06 54 567	51 78 999	Marble	Protomylonite			×		
Naxos high-grade core	8	M-8-4	06 54 567	51 78 999	Marble	Protolith			×	
		M-8-5	06 54 567	51 78 999	Marble	Protolith			×	
		8a1a	37°05'36.4"	025°28'53.0"	Marble	Mylonite	×	×	×	×
		8a1b	37°05'36.4"	025°28'53.0"	Marble	Protomylonite			×	
		8a1c	37°05'36.4"	025°28'53.0"	Marble	Protolith		×	×	
	11	11a1a	37°05'12.6"	025°28'20.0"	Marble	Mylonite	×	×	×	
		11a1b	37°05'12.6"	025°28'20.0"	Marble	Protomylonite			×	
		11a1c	37°05'12.6"	025°28'20.0"	Marble	Protolith		×	×	
		11a2	37°05'12.6"	025°28'20.0"	Marble	Mylonite				×
		11a3	37°05'12.6"	025°28'20.0"	Marble	Protolith	×			
		11b	37°05'12.6"	025°28'20.0"	Micaschist	–				

^a Map datums: UTM European Datum 1950 (Schneeberg Comble); WGS84 (Naxos high-grade core); ×Schneeberg Complex.

^b In distance to the late stage shear zones.

^c Several generations of syndeformational veins; thin section used for cathodoluminescence.

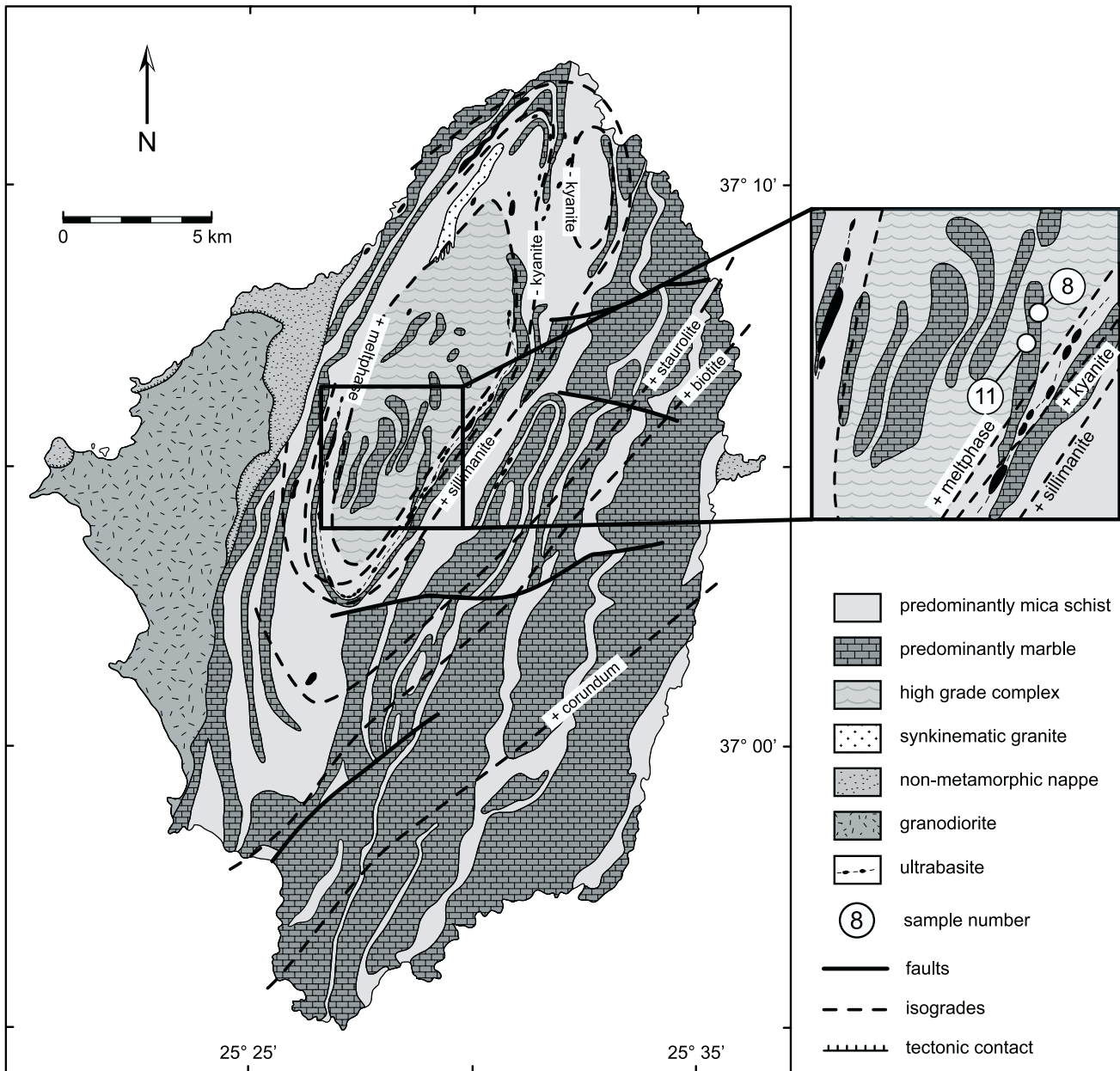


Fig. 2. Simplified geological map of Naxos (after Urai et al., 1990) with a detailed overview of the studied outcrops (coordinate system: WGS 84).

mylonites are white, porcelain-like layers alternating with yellowish bands of ferrous compounds formed by alteration of thin mica layers. Thin (mm scale) calcite veins inside the mylonites were observed in some outcrops.

The marble shear zones are up to 5 m thick and often have alternating layers with different degrees of recrystallization. The D_4 mylonitic lineation trends top towards WNW, i.e. the shear sense is sinistral, agreeing with Sölva et al. (2001).

The mica schists in the sampling area contain euhedral garnets with sizes up to 10 mm. Away from the shear zones, the garnets are brownish-red, but close to the shear zones the mica schists are often intercalated as 3–10-cm-thick layers with garnets often being greenish. The mylonitization is restricted to the weak calcite marble units.

3.1.2. Microstructures

Grains in the calcite marble host rocks are coarse (up to 2 mm) with lobate grain boundaries, suggesting dynamic recrystallization at high temperatures (Fig. 3a). The host rock is predominantly calcite, but also contains small amounts of randomly distributed second phases with a volume fraction of less than ~2%. Quartz grains are commonly rounded with sizes up to 100 μm , while muscovite occurs as flakes up to 200 μm in size.

Inside the shear zones, the protomylonites show the typical core and mantle structure. Judging from optical extinctions, the subgrains in the core structures have the same size as the fine mantle grains, suggesting that subgrain rotation recrystallization was the dominant recrystallization

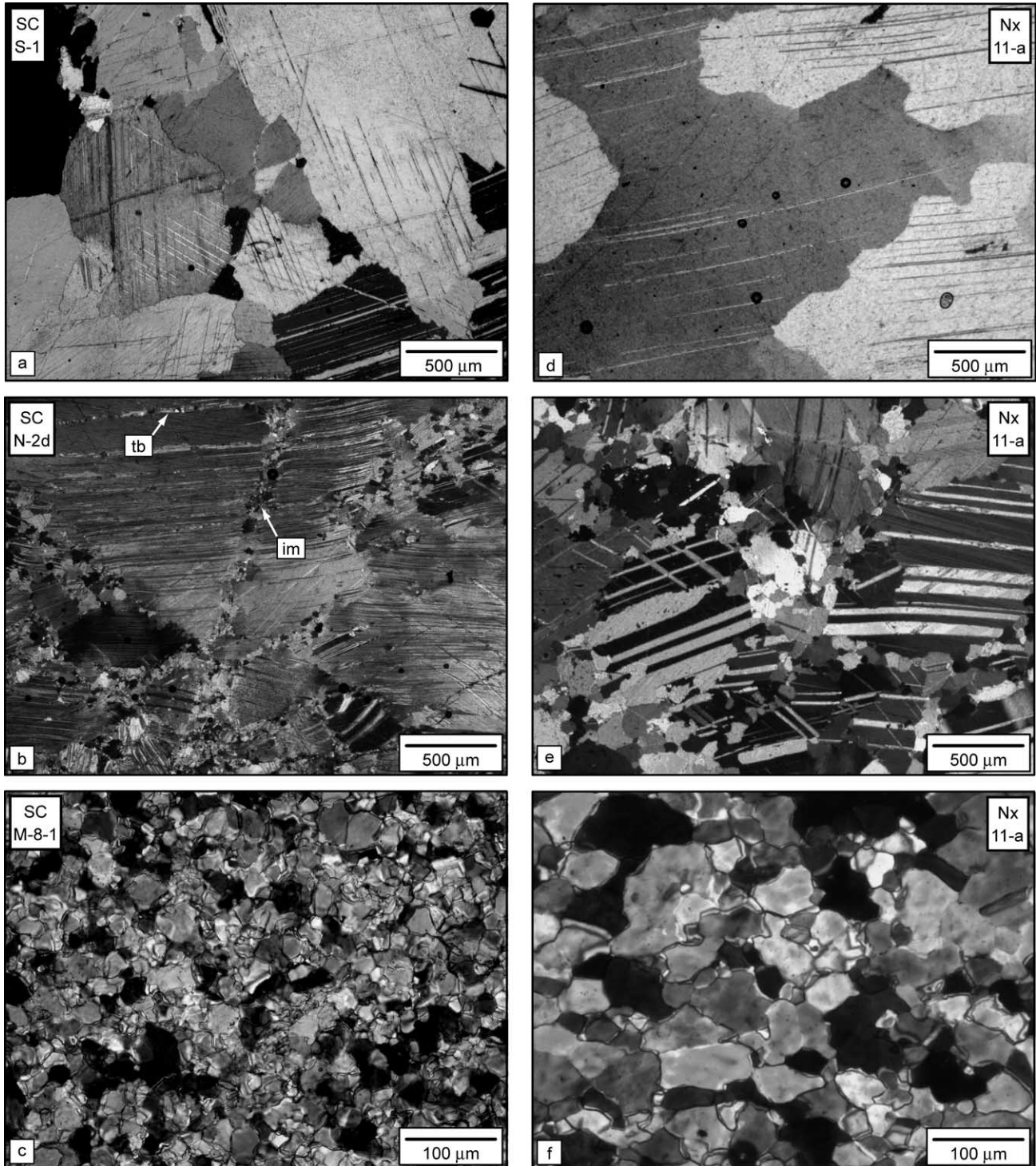


Fig. 3. Optical micrographs (transmitted light, crossed nicols) of marble samples from the Schneeberg area ((a)–(c)) and the Naxos high-grade core ((d)–(f)). (a) Coarse-grained, dynamically recrystallized marble host rock in the Schneeberg complex, not affected by late-stage D_4 shear zones. (b) Typical D_4 -protomylonite of the Schneeberg Complex; subsequent recrystallization by subgrain rotation results in the core and mantle structure; recrystallization commonly starts at twin boundaries (tb); in addition, the coarse, old grains are often cut by intragranular microcracks (im). (c) Typical mylonitic microstructure due to complete recrystallization during strain localization. (d) Dynamically recrystallized, coarse-grained, marble host rock inside the high-grade core of Naxos, with subgrain rotation recrystallization as dominant recrystallization process and thin twins being slightly curved. (e) Protomylonite with the typical core and mantle structure, presumably resulting from subgrain rotation recrystallization. (f) Mylonitic microstructure due to complete recrystallization during strain localization.

process. The coarse grains are characterized by undulatory extinction and kinking, both indications of strong plastic deformation. The presence of thick curved twins (type III after Burkhard, 1993) inside the coarse grains, and the fact that twin boundaries appear to have migrated suggests that mylonitization temperatures were higher than 250 °C (Burkhard, 1993; Ferrill et al., 2004). The coarse grains are often cut by linear arrays of fine grains. These features are likely intragranular microcracks that have, in turn, been recrystallized. In addition, recrystallized grains are often found along twin boundaries, forming intragranular shear zones (Fig. 3b). Within the mylonite zones, the marbles are completely recrystallized (Fig. 3c) with final grain sizes of 5–20 µm. The grain size data is derived by measuring the equidimensional circular diameter on polished and etched surfaces (see Herwegh, 2000). Applying the mean square root grain size of 10 µm to Rutter's (1995) sub-grain rotation piezometer differential stresses of 107 MPa are

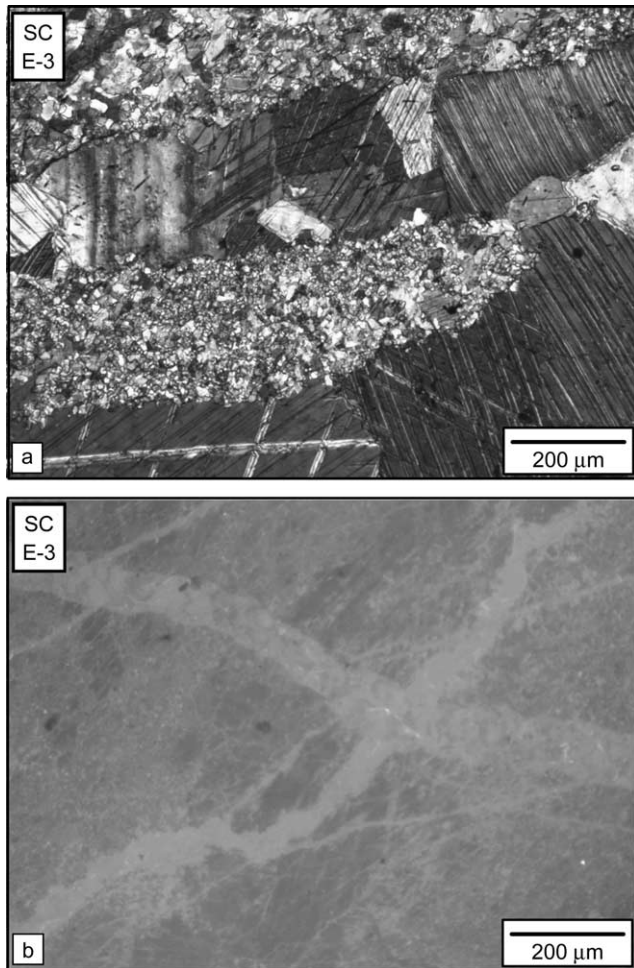


Fig. 4. Syndeformational calcite veins inside the D₄ marble mylonites. (a) Optical micrograph (transmitted light, crossed nicols). (b) Cathodoluminescence micrograph showing different generations of calcite veins; the color of the veins is brighter than that of the fine-grained marble mylonite matrix, probably owing to Mn²⁺ substitution and suggesting that the fluids were externally derived.

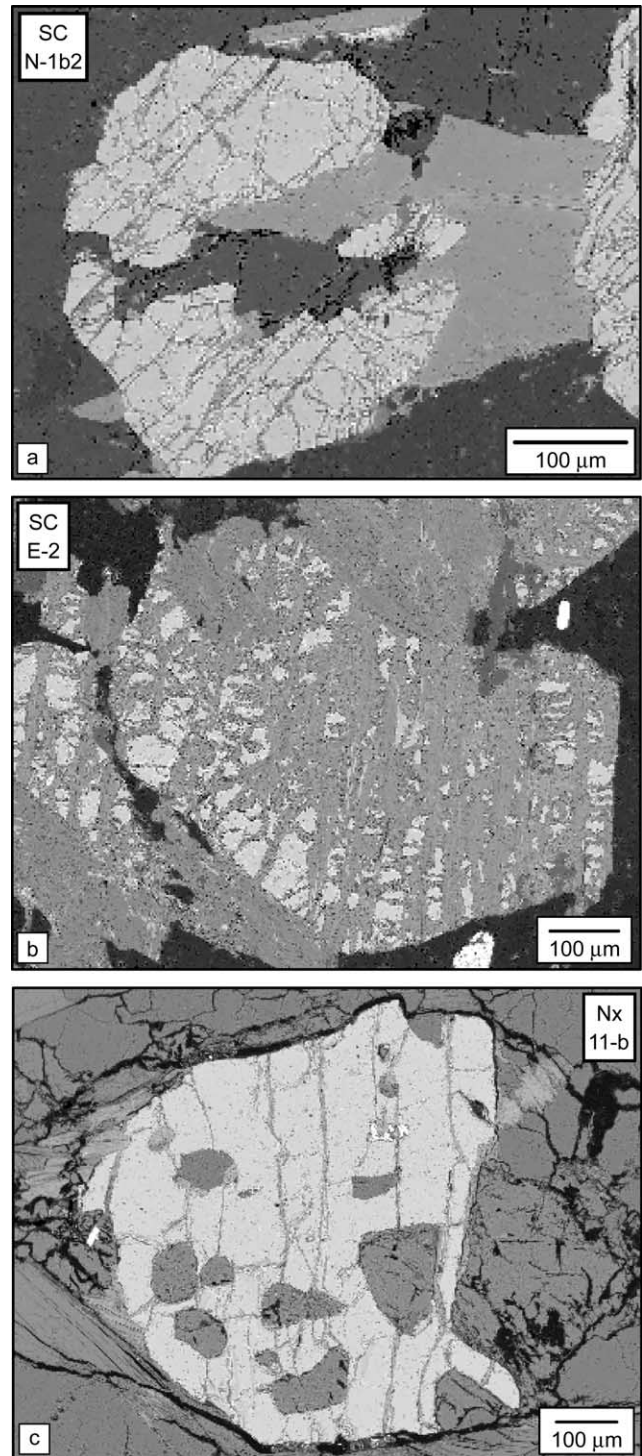


Fig. 5. Backscattered electron images of garnets in mica schists from the Schneeberg area ((a) and (b)) and from the Naxos high-grade core (c). (a) Garnet in a mica schist located in the distance of the D₄ shear zones in the Schneeberg area showing no evidence of alteration to chlorite or any other retrograde reaction of garnet or mica. (b) Garnet in a mica schist located just next to a D₄ marble shear zone in the Schneeberg complex; the garnet's original shape is still visible, but it is highly altered to chlorite by retrograde reactions due to the involvement of fluids. (c) Garnet inside mica schist just next to a marble mylonite inside the high-grade core of Naxos; the garnet is not affected by retrograde reactions and its appearance is similar to the garnets sampled in the distance of the late-stage shear zones.

calculated. In some outcrops, the mylonites contain calcite veins (Fig. 4a) that are deformed and recrystallized again.

The garnet-mica schists of the Laaser Series consist of quartz, mica, feldspar and almandine-rich garnet. Away from the D₄ zones, backscattered electron micrographs of the garnets do not show any alteration (Fig. 5a). However, close to the marble shear zones the garnets are highly altered to chlorite pods that preserve the garnet's original shape (Fig. 5b). Other retrograde reactions are common, including sericitization of plagioclase pointing to the activity of fluids under lower greenschist facies.

Hot cathodoluminescence (CL) was used to obtain qualitative information on the chemical distribution inside the Schneeberg marble mylonites. Whereas the fine-grained matrix is characterized by a dull (brown-orange) luminescence, the veins can be distinguished by a brighter color (yellow-orange) (Fig. 4b). The bright color is due to substitution of the Ca²⁺ site by Mn²⁺ (Machel and Burton, 1991; Lewis et al., 1998; Barbin, 2000) and suggests the presence of externally derived fluids with a different chemical composition (open system). Crosscutting relationships of the veins and different degrees of diffusion of the luminescence intensity point to several generations of fracturing and crystallization (fracture-sealing) during mylonitization.

To study the grain boundary morphology of the recrystallized calcite and to minimize the influence of possible late-stage fluid infiltration on the grain boundary morphology, the samples were taken at a reasonable distance from the surface (tens of cm). The sections with a thickness of ~2 mm were broken after several cycles of heating (~220 °C) and cooling (~25 °C). The temperature cycling promoted intergranular fractures. The samples were sputtered with Au–Pd and observed using SEM.

Most grain boundaries of the Schneeberg mylonites are characterized by isolated, triangular pores. They differ in size, but are similar in shape and orientation, indicating that they are crystallography controlled (Fig. 6a–c). There is also evidence of a connected network of triple-junction tubes, with the dihedral angles being controlled by crystallographic orientation of the respective grains (Fig. 6c).

3.1.3. Chemistry

The chemical composition of the mylonites and their respective host rocks was analyzed by XRF and ICP-OES to investigate the influence of fluids during shear zone evolution (Tables 2 and 3). Additionally, EDX analysis on polished and etched surfaces and roentgen diffractometry on insoluble residues of the dissolved marble samples were used to gain additional information on the chemistry of those mineral phases.

The marble host rock of the Laaser Series is very pure (Tables 2 and 3). RDA and EDX analyses indicate muscovite and quartz being second phase minerals. Inside the shear zones the mylonites are enriched in some elements, especially Mn and Al (Fig. 7a). Some of the

elements can be attributed to the second phase minerals muscovite, biotite and chlorite. However, the ICP analysis shows that the calcite composition of the mylonites is commonly enriched in several other elements, including Na, Mg, and Ti (Fig. 7b). Inside the mylonites elements such as Mn and Mg are incorporated into the calcite lattice and are presumably derived from fluids that also promoted the alteration towards chlorite.

In addition to the chemical composition, the presence of fluids during shear zone evolution can be detected by measurements of stable isotopes. We selected rock chip samples of homogeneous marble with different degrees of recrystallization, traversing from the host rock to the center of the mylonite zone. The stable isotopes C¹³ and O¹⁸ were analyzed at the Mineralogical Department of the University of Bonn, Germany (R. Hoffbauer) (Table 4) and are displayed in PDB and plotted as a function of the degree of recrystallization (Fig. 8a). Whereas the respective host rock data range between –12 and –9‰ for δ¹⁸O and between 0.95 and 1.8‰ for δ¹³C, there is far greater scatter of values in the recrystallized parts, ranging from –14 to –6‰ for δ¹⁸O and from 0.3 to 1.6‰ for δ¹³C. In addition, isotope data derived from a syndeformational vein that formed inside a mylonite (E-3; with values of δ¹⁸O = –13.69‰ and δ¹³C = –0.42‰) indicates that the fluids were derived from an external source.

To constrain the temperature at which D₄-mylonitization took place, calcite–dolomite solvus geothermometry was applied. According to Matthews et al. (1999), calcite–dolomite geothermometry can be applied in recrystallized calcite (due to cation equilibration) even at temperatures below 400 °C. The analyses for Ca and Mg were made using an electron microprobe (Table 5). The temperatures, calculated according to the equation of Lieberman and Rice (1986) indicate mylonitization temperatures of 279 ± 25 °C, in agreement with the presence of type III calcite twins, the existence of twin boundary migration and with the studies of Sölvä et al. (2005) who proposed lower greenschist facies conditions for the D₄-Laaser Series shear zone.

3.2. Naxos high-grade core

3.2.1. Mesostructures

Inside the high-grade core of Naxos, the marble rafts are predominantly of calcite with rare quartz grains. The calcite marble, famous for its purity and white color and mined since ancient times, is very coarse-grained with grain sizes up to 15 mm due to peak *M*_{2b} conditions (~700 °C and ~0.6 GPa). The marble rafts are intercalated by pegmatite intrusions and layers of amphibolite and mica schist. Mylonite zones are restricted to the marble units and are easily detected owing to the striking difference in grain size and the milky, porcelain-like appearance. The shear zones have a thickness of up to 1 m with alternating degrees of

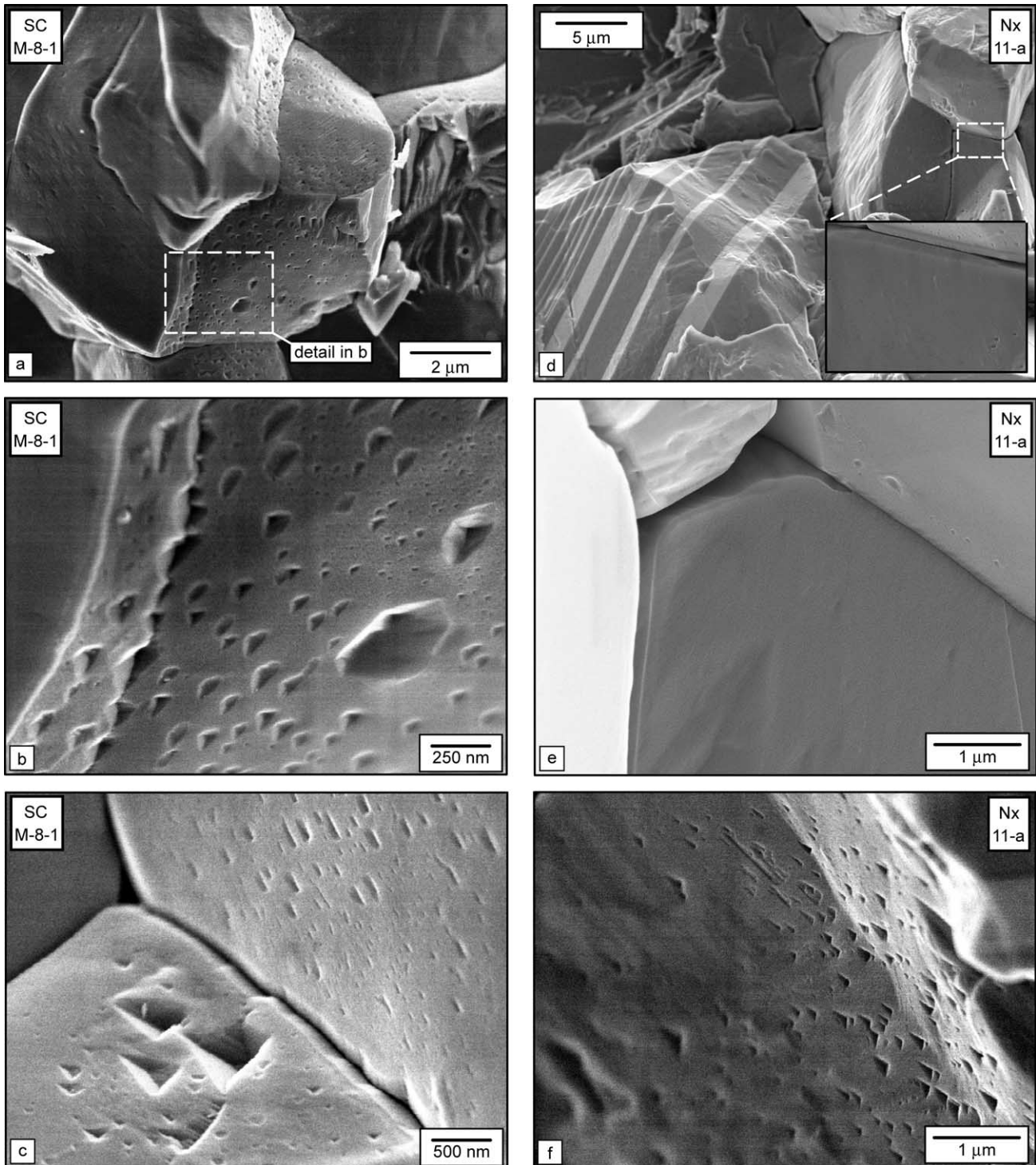


Fig. 6. SEM micrographs of broken surfaces of marble mylonites from the Schneeberg Complex ((a)–(c)) and from the high-grade core in Naxos ((d)–(f)) showing the grain boundary morphology. (a) and (b) Grain boundaries contain triangular pores controlled by crystallography. (c) The pores differ in volume, but their orientation and shape is similar; note also the tubes along triple grain junctions and the dihedral angle. (d) In the Naxos rocks, grain boundaries are smoother and contain only a few small pores; note the 3D topography of the heavily thick twinned grain in the lower left corner. (e) Rarely, triple junctions contain tubular pores. (f) In a few cases, isolated, crystallography controlled pores are observed on a grain boundary; suggesting that a small amount of fluids was present in the Naxos mylonites.

recrystallization; the stretching lineations trend N–S (Urai et al., 1990).

The intercalated layers of mica schist and amphibolite

are up to 10 cm thick. Due to the complex deformation history that includes N–S extension during D_2 , the layers are boudinaged and folded with fold axes trending N–S.

Table 2
XRF data for some elements of selected samples (values in wt%)

	Relative accuracy (%)	Schneeberg complex								Naxos high-grade core		
		N-1b1	N-2 h	E-1-1	E-1-2	E-4	M-8-1	M-8-3	M-8-4	8a	11a-1	11a-3
Fe ₂ O ₃ (T)	0.39	0.19	0.12	0.12	0.07	0.14	0.22	0.16	0.12	0.02	0.02	0.03
TiO ₂	0.03	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Al ₂ O ₃	0.24	0.01	0.11	0.40	0.12	0.09	0.17	0.12	0.15	0.33	0.06	0.04
MnO	0.02	0.01	0.02	0.01	0.01	0.02	0.17	0.08	0.03	0.01	0.01	0.01
MgO	0.16	0.26	0.45	0.33	0.28	0.28	0.14	0.32	0.56	0.36	0.48	0.44
Na ₂ O	0.08	0.14	0.18	0.18	0.25	0.17	0.18	0.13	0.32	0.29	0.19	0.14

The mica schists contain a small number of euhedral garnets. Their grain size is up to 2 mm, and they are reddish-brown, both near to and far from the shear zones.

3.2.2. Microstructures

The marble far from the shear zones is coarse-grained with lobate grain boundaries, pointing to high-temperature dynamic recrystallization (at peak M_{2b} conditions; Urai et al., 1990) (Fig. 3d). As in the Schneeberg Complex, the Naxos protomylonites are also characterized by core and mantle structures, indicating subgrain rotation recrystallization (Fig. 3e). In contrast to the Schneeberg samples, the Naxos protomylonites do not contain intragranular microcracks or shear zones.

Subsequent strain localization is accompanied by complete mylonitization to grain sizes ranging between 20 and 50 μm (Fig. 3f). A final grain size of 25 μm (mean square root) corresponds to a stress of 48 MPa, using the equation for rotation recrystallization of Rutter (1995). We did not observe any veins inside the Naxos mylonites.

Similar to the mica schists of the Laaser Series in the Italian Alps, the intercalated mica schists inside the marble

rafts of the Naxos high-grade core consist of quartz, mica, feldspar and almandine, but have lower garnet content. Backscattered images of garnets in mica schists, both far from and near to the shear zones are very similar and do not show any chloritization (alteration) of the garnets or sericitization of plagioclase (Fig. 5c). Thus, we infer that the mica schists were not affected by late-stage retrograde metamorphic reactions.

Most grain boundaries of the Naxos mylonites are slightly curved and occasionally contain pores (Fig. 6d). Minor porosity is present at triple junctions, rarely within triple junction tubes (Fig. 6e). Uncommonly, isolated crystallography-controlled pores are present on the grain boundaries of these samples (Fig. 6f) similar in appearance to those observed in the Schneeberg.

3.2.3. Chemistry

As in the Schneeberg marbles, the Naxos high-grade marble is characterized by its purity (Tables 2 and 3). RDA and EDX analyses show that quartz and muscovite together with very rare amounts of feldspar are present as randomly distributed second phases with contents of less than 0.5%.

Table 3
ICP-OES data for some elements of selected samples (values in ppm)

	Schneeberg complex			Naxos high-grade core			
	E			8a		11a	
	h.r. ^a	myl. ^b	myl.	h.r.	myl.	h.r.	myl.
	E-1-4	E-1-1	E-4	8a1c	8a1a	11a1c	11a1a
Mn	24.0	46.2	125.2	11.5	8.6	6.6	20.4
Fe	320.0	493.0	665.3	22.2	130.5	34.6	40.2
Ni	0.2	0.3	0.4	0.3	0.1	0.7	0.2
Cu	0.1	0.2	0.1	0.2	0.1	0.2	0.3
Zn	0.6	1.5	0.8	0.3	0.4	0.7	1.1
Cr	0.0	0.2	0.0	0.4	0.4	0.2	0.1
Ti	1.0	2.1	1.1	1.6	1.3	1.1	1.0
Mg	1343.5	1087.5	1190.3	1421.8	1209.1	2063.9	1558.4
Sr	96.4	96.1	384.7	66.5	52.1	80.8	69.7
Al	13.5	24.9	26.8	1.6	1.5	15.6	2.2
Si	14.9	20.9	21.3	3.3	2.8	4.2	4.2
Na	28.6	19.9	20.2	10.0	10.4	11.2	17.2
K	7.2	6.2	7.9	2.3	2.0	3.3	5.1
P	9.9	26.9	29.8	24.6	26.9	41.9	24.3

^a Host rock.

^b Mylonite.

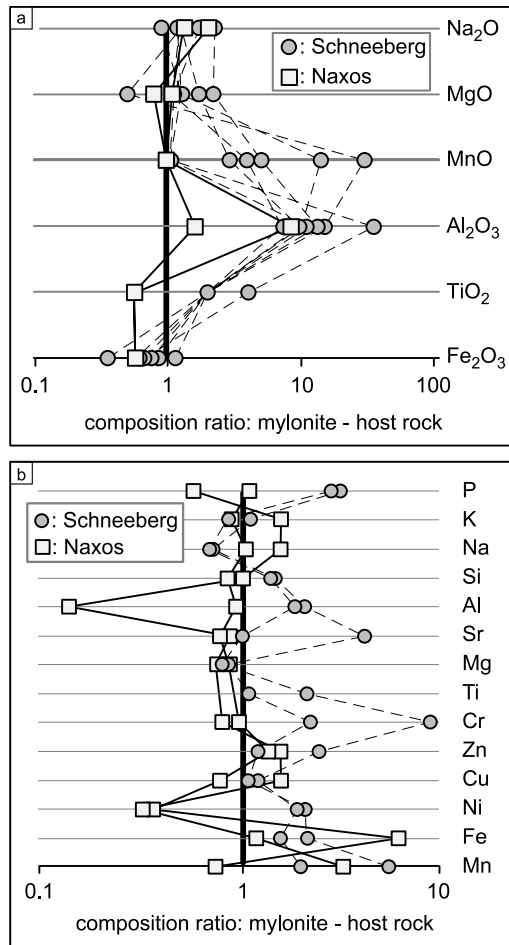


Fig. 7. Plots of chemical analyses of the selected mylonites normalized to their respective host rocks derived by (a) XRF and (b) ICP-OES (circles: Schneeberg Complex; squares: Naxos high-grade core; thick line at one: host rock composition). The Schneeberg mylonites show a larger variation in some elements compared with their respective host rocks, whereas the Naxos mylonites do not show these significant compositional changes.

The quartz grains are rounded and are characterized by sizes of commonly 10 μm . The two mylonites are very similar in chemical composition compared with their host rock (Fig. 7a and b). However, compared with the Schneeberg shear zones, the chemical composition of the Naxos mylonites deviates only slightly from that in the respective host rock.

We analyzed carbon and oxygen isotopes along cross-sections of two different shear zones inside the Naxos high-grade core (Table 4). The host rock data is about 2.1‰ for $\delta^{13}\text{C}$ and ranges between -8 and -5.5 ‰ for $\delta^{18}\text{O}$, the latter being in accordance with data from Baker and Matthews (1995). The isotopic signature of the recrystallized mylonites is very similar to the host rock (Fig. 8b). Compared with the Schneeberg mylonites, the Naxos samples show much smaller changes in composition. Applying the calcite–dolomite solvus geothermometry for the post- M_{2b} mylonites inside the high-grade core of Naxos indicates temperatures of 271 ± 15 °C (Table 5), i.e. close to those calculated for the Schneeberg mylonites.

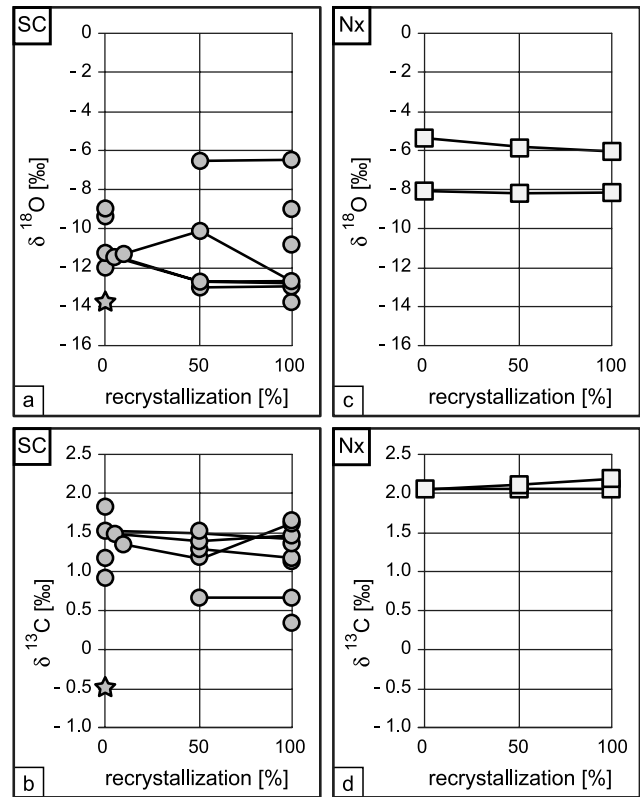


Fig. 8. Diagrams of stable isotopes C^{13} and O^{18} (both: PDB standard) as cross-sections from host rock (0% RX) towards mylonite (100% RX) for several shear zones in the Schneeberg Complex ((a) and (b)) and the Naxos high-grade core ((c) and (d)). The recrystallized parts in the Schneeberg marble shear zones show a large scatter of data for both C^{13} and O^{18} , whereas there are only minor changes throughout the profiles in the Naxos marble shear zones. The asterisk represents the vein data inside a Schneeberg marble mylonite (E-3).

4. Discussion

The host rocks and the marble mylonites from the two study areas are very similar. Both of the host formations consist of calcite marbles of high purity, intercalated between mica schist, and both have an early history of polyphase, high-grade metamorphism, accompanied by deformation that produced massive, coarse-grained rocks. Both hosts suffered a second deformation episode that took place during uplift, and that resulted in localized deformation in shear zones, at temperatures around 270–280 °C.

The shear zones in the two study areas are similar, too: strain localization resulted in progressive recrystallization of the coarse-grained host into a fine-grained marble mylonite. Intermediate stage protomylonites have core and mantle structures evolving by a combination of subgrain rotation and grain boundary migration. The recrystallized grain size of the mylonites is slightly different, indicating higher differential stress in Schneeberg (~ 100 MPa) than Naxos (~ 50 MPa). If the two marbles had the same rheology, the difference in grain size would suggest that strain rates in Schneeberg were higher, but experiments

Table 4
Data of the analyses of the stable isotopes C¹³ and O¹⁸ displayed in PDB standard

	Samples	% (Recr.)	δ ¹⁸ O (‰) (SMOW)	δ ¹⁸ O (‰) (PDB)	δ ¹³ C (‰) (PDB)
Schneeberg complex	M-8-1a	100	18.42	−12.98	1.18
	M-8-1b	50	18.46	−12.94	1.30
	M-8-2a	100	18.53	−12.87	1.39
	M-8-2b	50	18.64	−12.76	1.53
	M-8-3a	50	18.65	−12.75	1.48
	M-8-3b	100	18.66	−12.74	1.45
	M-8-3c	50	18.66	−12.74	1.44
	M-8-4	5	19.97	−11.47	1.51
	M-8-5	0	20.19	−11.26	1.53
	E-1-1	100	18.56	−12.84	1.62
	E-1-2	50	21.35	−10.13	1.23
	E-1-3	10	20.09	−11.36	1.36
	E-3	0 (vein)	17.69	−13.69	−0.42
	E-4	100	17.64	−13.73	0.34
	N-2d1	100	22.51	−9.01	1.67
	N-2h	100	20.65	−10.81	1.16
	N-2a2a	100	25.08	−6.51	0.63
	N-2a2b	50	25.03	−6.56	0.67
	N-1a1	0	22.49	−9.03	1.81
	N-1b1	0	22.13	−9.37	1.21
N-1c1	0	19.42	−12.01	0.95	
Naxos high-grade core	8a1a	100	23.42	−8.12	2.06
	8a1b	50	23.38	−8.16	2.06
	8a1c	0	23.40	−8.14	2.07
	11a1a	100	25.65	−5.96	2.19
	11a1b	50	25.80	−5.81	2.09
	11a1c	0	26.15	−5.47	2.06

Sample E-3 represents data of a syndeformational vein.

indicate that there are large differences in rheology of calcite marbles of only slightly varying composition (e.g. de Bresser et al., 2002).

The main difference between the two study areas is the fugacity of water in the fluids present during mylonite formation. The role of CO₂ within the pore fluid must not be neglected, as calcite needs CO₂ to be stable. Clearly, detailed knowledge of the fluid chemistry would contribute to a better understanding of the mass transfer processes that were active during shear zone evolution. From our present data, we propose that the difference of the presence of fluids, and the fact that most other parameters are similar, forms the basis for evaluating the role of fluids in calcite shear zones at temperatures around 270–280 °C. A schematic model summarizing the discussion below is shown in Fig. 9.

We infer that the Schneeberg D₄ mylonites were formed in the presence of an abundance of aqueous fluids based on the following: the calcite composition of the mylonites was altered during shear zone evolution; mica flakes within these rocks were altered to chlorite; stable isotopes within the mylonites were depleted with respect to their host rocks; and finally chemical changes were suggested by CL observations. In addition, syndeformational calcite veins and the retrograde garnets in adjacent mica schists provide additional evidence for fluids. A late syndeformational vein in the Schneeberg mylonite has the isotopic signature of a metamorphic fluid of an external source.

These changes in mineralization, solid–solution impurities, and isotope concentrations require advective transport by a fluid, combined with local redistribution on the grain

Table 5
Microprobe data of several marble mylonites used to calculate the temperature during mylonitization from calcite–dolomite solvus geothermometry (Lieberman and Rice, 1986). The entire data set is available from the authors upon request.

	Sample	n ^a	wt% CaO		wt% MgO		X _{MgCO₃}		T (°C) ^b	
			Mean	Std dev	Mean	Std dev	Mean	Std dev	Mean	Std dev
Schneeberg	N-2h	23	55.01	0.4104	0.39	0.1106	0.0083	0.00234	300	24
	E-1-1	27	55.55	0.2839	0.25	0.0400	0.0052	0.00084	262	13
	E-3	8	54.33	0.3066	0.29	0.0443	0.0062	0.00096	276	13
Naxos	8a1a	20	55.61	0.3287	0.23	0.0235	0.0049	0.00049	258	8
	11a2	19	54.30	0.3418	0.32	0.0251	0.0068	0.00052	285	6

^a Number of measurements.

^b Calculated according to the equation of Lieberman and Rice (1986) (T (°C) = $3685.7 / (1.6145 - \ln X_{MgCO_3}) - 273$).

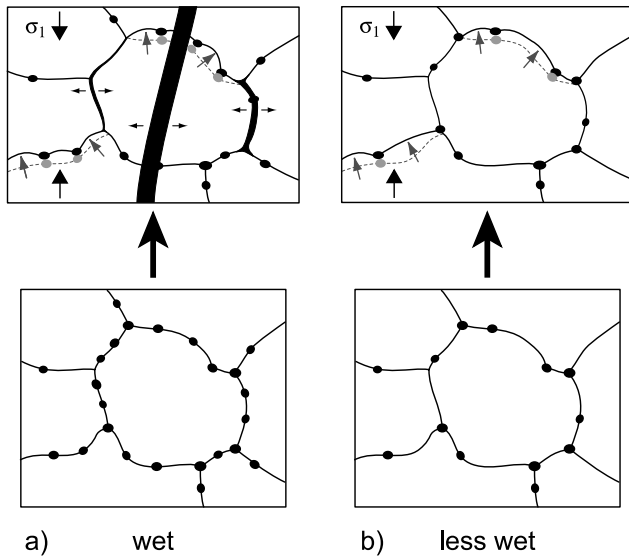


Fig. 9. Sketches of the microstructures of the marble mylonites recrystallized under (a) wet and (b) less wet conditions. The pre-mylonitic structure, sketched in the lower two diagrams, is assumed to be similar in both regimes, except for the amount of fluids present as pores on the grain boundaries. During shear zone evolution, veins, intragranular cracks and dilatant grain boundaries, result from the activity of fluids inside the wet system; features that are absent, or at least much less numerous in the drier (less-wet) system. However, in both systems, any porosity that is present is probably dragged by slow grain boundary migration during mylonitization (indicated by small arrows), resulting in similar grain boundary morphology (as observed in SEM).

scale. The details of this process are apparently complex: in most mylonite samples calcite is enriched in e.g. manganese and titanium as shown by ICP data. The incorporation of Mn into the calcite lattice can occur during recrystallization (Olgaard, 1985; McCaig et al., 1999). Similar Mn enrichments in marble mylonites, documented by CL, have led other authors to argue for the presence of externally derived fluids (Busch and Van der Pluijm, 1995; Badertscher and Burkhard, 2000). Other studies (e.g. Bestmann et al., 2000) document the formation of mylonite without pervasive fluid flow.

In contrast to the Schneeberg mylonites, the Naxos mylonites do not show significant differences with respect to the host rocks either in bulk chemical composition or in stable isotopes. The Naxos marbles do not contain calcite veins and the garnets in the adjacent mica schists were not affected by retrograde reactions. The lack of differences between host and mylonite suggests that the Naxos mylonites were formed under much less 'wet' conditions than the Schneeberg mylonites.

Because of leakage during uplift (associated with grain boundary cracking owing to the high thermal expansion anisotropy of calcite) we do not know what the speciation of the fluid in the grain boundary pores was during recrystallization. Based on the chemical changes and pore morphology we interpret these to have been fluid-filled. Such triangular pores have been observed during crack-

healing in single-crystal calcite when water is present (Hickman and Evans, 1987) and on grain boundaries in Cararra marble annealed with water at 800 °C (see fig. 5e in de Bresser et al., 2005). Such experiments are, of course, done under conditions at which late-stage fluid infiltration (e.g. rain water) can be excluded.

We now ask the question how the presence of fluids in the Schneeberg D₄ shear zones has influenced microstructural evolution. Multiple generations of dynamically recrystallized calcite veins indicate that the veins were formed and deformed during mylonitization, probably by periodic influx of high-pressure fluid with subsequent precipitation of calcite, as the rocks were exhumed along the Schneeberg Normal Fault Zone (SNFZ). Therefore, the changes in calcite composition may have been caused by two processes: (1) new calcite crystallized from the fluid and then subsequently recrystallized (see also Kennedy and Logan, 1997; Badertscher et al., 2002; Herwegh and Kunze, 2002), or (2) impurities were incorporated during dynamic grain boundary migration recrystallization. In fact, recrystallized intragranular fractures inside the coarse grains point to the combined activity of these processes. Grain boundaries containing many isolated pores and connected tubes along triple grain junctions are also apparently associated with recrystallization processes. Under conditions of low effective pressure, grain boundaries may have dilated and formed new porosities, some of which may have subsequently been removed by compaction when the fluid pressure dropped.

In the Naxos mylonites, most of the processes described above are absent. Triple junction porosity exists, but such porosity is most commonly isolated, that is, unconnected. The permeability of these mylonites is probably very low. Rare grain boundaries with many pores may have been associated with local fluid enrichment. Such pores probably hindered grain boundary migration by Zener drag, particularly if they were filled with a non-aqueous fluid (Fig. 9) (Stüwe, 1978; Olgaard, 1985; Urai et al., 1986a; Evans et al., 2001; Herwegh and Kunze, 2002; Herwegh and Berger, 2004).

Returning to the Schneeberg mylonites, it is then interesting to note that the presence of fluids did not have a major effect on grain boundary morphology and recrystallization behavior. Although the amount of pores on grain boundaries is different, other morphological characteristics are similar. The recrystallization mechanisms are similar, too: subgrain rotation recrystallization followed by slow grain boundary migration. Thus we infer that drag of the grain boundary fluid in pores occurred in the Schneeberg mylonites as well.

5. Conclusions

To assess the effect of water-rich fluids on the recrystallization behavior and grain boundary morphology

in natural calcite, we compared marble mylonites from the Schneeberg Complex in the Italian/Austrian Alps to mylonites from the high-grade core of Naxos, Greece. Both settings have similar geologic histories, but they differed in the nature of the fluids present during mylonitization. Both the fluid-rich (Schneeberg) and fluid-poor (Naxos) marble mylonites have similar grain boundary microstructures. The microstructural evolution inside the Schneeberg mylonites was affected by the presence of fluids as shown by (1) the introduction of syndeformational calcite veins with slightly different chemical composition, (2) the presence of intragranular cracks inside the protomylonites, (3) evidence for dilatant grain boundaries, and (4) the existence of triple junction tube porosity inside the mylonites. None of these features are present in the Naxos samples. However, this presence of fluids apparently did not affect the recrystallization behavior, at least as can be judged by the microstructure: The dominant recrystallization process of both types of mylonites was probably subgrain rotation recrystallization followed by slow grain boundary migration. In addition the grain boundary morphology is roughly similar, even if the amount of porosity residing on the grain boundaries is different.

Apparently, the fluids did not have a major influence on recrystallization behavior and grain boundary morphology in calcite, at least for these two marble mylonites.

Such a conclusion is in agreement with deformation experiments of Carrara marble at high temperatures and high strain rates (de Bresser et al., 2005), but is in strong contrast to inferences concerning the effect of water on recrystallization and grain boundary migration and morphology in other minerals such as quartz, halite, olivine or feldspar.

Acknowledgements

The authors would like to thank M. Burkhard and M. Herwegh for the critical and constructive reviews and comments. We are grateful to R. Hoffbauer for carrying out the stable isotope measurements. A. Heimann is thanked for his assistance with sample preparation and analytical work. M. Frongillo, J. Kallinna and U. Wollenberg are thanked for their help with the SEM. We also thank N. Chatterjee for his essential assistance with the microprobe and S. Sintern and R. Neef for carrying out the ICP-OES analyses. We acknowledge W. Kraus for preparing thin sections, H. Sólva for providing an early version of his paper and H. de Bresser for his valuable comments on the recrystallization of marble. JLU thanks M.I. Spalla for discussions on Schneeberg geology. OS acknowledges Prof. Dr. K. Heitfeld Stiftung for the financial support during his stay at MIT. This project is funded by the Deutsche Forschungsgemeinschaft (UR 64/4-1). BE gratefully acknowledges funding from NSF EAR 0309510.

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