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Grain coarsening in polymineralic contact metamorphic carbonate rocks: The role of different physical interactions during coarsening

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ABSTRACT

The microstructural evolution of polymineralic contact metamorphic calcite marbles (Adamello contact aureole) with variable volume fractions of second-phase minerals were quantitatively analyzed in terms of changes in grain size and nearest neighbor relations, as well as the volume fractions, dispersion and occurrences of the second phases as a function of changing metamorphic conditions. In all samples, the calcite grain size is controlled by pinning of grain boundaries by second phases, which can be expressed by the Zener parameter (Z), i.e., the ratio between size and volume fraction of the second phases. With increasing peak metamorphic temperature, both the sizes of matrix grains and second phases increase in dependence on the second-phase volume fraction. Two distinct coarsening trends are revealed: trend I with coupled grain coarsening limited by the growth of the second phases is either characterized by large-sized or a large number of closely spaced-second phase particles, and results finally in a dramatic increase in the calcite grain size with Z. Trend II is manifest by matrix controlled grain growth, which is retarded by the presence of single second-phase particles that are located on calcite grain boundaries. It is supported by grain boundary pinning induced by triple junctions, and the calcite grain size increases moderately with Z. The two different grain coarsening trends manifest the transition between relatively pure polymineralic aggregates (trend II) and microstructures with considerable second-phase volume fractions of up to 0.5. The variations might be of general validity for any polymineralic rock, which undergoes grain coarsening during metamorphism. The new findings are important for a better understanding of the initiation of strain localization based on the activation of grain size dependent deformation mechanisms.

1. Introduction

The growth of minerals is a fundamental process in all rocks that underwent metamorphism (e.g., Joesten, 1991). Knowledge on the changes in grain size is important, because grain size increases are directly related to changes in the grain boundary network affecting the mass-transfer pathways on the grain-scale, which is relevant for many metamorphic and deformed rocks in the Earth's crust and mantle. Under static conditions, a monomineralic rock increases its grain size continuously, following grain growth kinetics as defined by exponential laws (Simpson et al., 1971; Poirier and Guillopé, 1979; Joesten, 1983, 1991; Olgaard and Evans, 1986). Surface energy is the main driving force for grain growth in such pure aggregates (Evans et al., 2001 and references therein), which might be counteracted, however, by pinning of grain boundaries due to triple junction drag (Shvindlerman and Gottstein, 2005; Piazolo et al., 2006).

In nature, rocks are rarely monomineralic and very small volume fractions of additional minerals, so-called second phases, impose an effect on grain growth of the matrix phase (Riege et al., 1999; Weygand et al., 2000; Berger and Herwegh, 2004; Herwegh and Berger, 2004; Ferry et al., 2005; Zheng et al., 2006). Generally speaking, the mean grain size in a polymineralic aggregate is the time integrated product of the grain coarsening process at elevated temperatures as is the case for contact metamorphism, for example. In order to understand grain coarsening and to predict the evolution of grain sizes in such polymineralic rocks, an understanding of the effect of the second phases on the migrating grain boundaries of the matrix phase is crucial. In this sense, second phases can counteract the surface energy of matrix grains by imposing a dragging force on the migrating grain boundaries, resulting in a retardation of grain growth of the matrix phase (see Fig. 6, Brodhag and Herwegh, 2009) or even in a complete immobilization of the grain boundaries inducing a stabilization of the matrix's grain size. Hence, the second phases can directly control the grain size of the matrix phase. Generally, second phases have to be

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List of symbols and abbreviations

c	constant in the Zener equation
cc	calcite
CPP	clustered particle pinning
D	mean matrix grain size
D_{cc}	mean calcite matrix grain size
$D_{\max}$	maximum matrix grain size
D_{nor}	mean norcamphor matrix grain size
d_p	mean second-phase grain size
f_p	second-phase volume fraction
gb	grain boundary (of a matrix grain)
K	nearest neighbor correlation parameter
m	parameter assigning the spatial location of second phases
M_{cc}	modes of nearest neighbor distribution of calcites
MPP	multiple particle pinning
M_{sp}	modes of nearest neighbor distribution of second phases
n(cc)	number of calcite grains
n(sp)	number of second-phase particles
sp	second phase
SPP	single particle pinning
t	time
T	temperature
TJP	triple junction pinning
Z	Zener parameter

discriminated into solid and liquid/gaseous second phases as they behave in a rigid and deformable manner respectively during pinning, imposing therefore different physical interactions onto a migration grain boundary (Evans et al., 2001; Petrishcheva and Renner, 2005). For both types the amount, size and dispersion of the second phases define if and how the grain boundaries of a matrix phase are affected. In the case of solid second phases, for example, a large number of nano-scale second phases is required (Herwegh and Kunze, 2002; Berger and Herwegh, 2004; Burhan and Ferry, 2006), while for large second phases (with grain sizes comparable to those of the matrix grains) only a small number of second phases are already sufficient to efficiently control the evolution of the grains of the matrix phase. It is this relation between size (d_p) and volume fraction (f_p) on the one side and the grain size of the matrix phase (D) on the other side, which was discovered by Zener (in Smith, 1948).

$$D = c \cdot \frac{d_p}{f_p} \tag{1}$$

This relation has meanwhile variously been modified to express the effect of second phases in polymineralic materials in the case of material sciences (Gladman, 1966; Hellman and Hillert, 1975; Evans et al., 2001; Brodhag and Herwegh, 2009) and for statically affected or deformed polymineralic rocks in nature (Mas and Crowley, 1996; Berger and Herwegh, 2004; Herwegh et al., 2005; Ebert et al., 2007a,b, 2008; Song and Ree, 2007; Herwegh et al., 2008). In addition to second phases, chemical impurities (e.g., Freund et al., 2001; Nakamura et al., 2005; Shvindlerman and Gottstein, 2005), melt films (Renner et al., 2002; Petrishcheva and Renner, 2005), crystallographic misorientations between adjacent grains (Stöckhert and Duyster, 1999; Kruhl, 2001; Bestmann and Prior, 2003; Wheeler et al., 2003) and grain boundary triple junctions (Pande and Masumura, 2005; Piazolo et al., 2005, 2006; Gottstein et al., 2006) can also affect grain growth in polymineralic rocks. Under the presence of solid state second phases, however, all these additional pinning parameters are of minor importance only, as suggested by Shvindlerman and Gottstein (2005) and experimental results of Brodhag and Herwegh (2009).

The effect of second phases on the growing matrix phase can be reduced to two general behaviors. (i) For low second-phase volume fractions, the matrix grain boundaries are pinned or dragged in the aforementioned manner but pinning by individual particles can be overcome as a function of time or increasing temperature. (ii) In the case of high second-phase volume fractions, a complete immobilization of the matrix grain boundaries by pinning due to the large amount of second-phase particles occurs. In this case, a grain size increase of the matrix phase can only take place when the second phases grow, giving more space to allow simultaneous growth of the matrix grains. This combined growth of matrix and second phase is referred to as coarsening and has been treated by experimental and numerical modeling approaches, where the latter either use the minimization in surface energy (e.g., Solomatov et al., 2002) or reduction in chemical gradients (e.g., Ardell, 1972) as major driving forces. In the case of polymineralic natural rocks, however, very little is known about the two effects of second-phase pinning since systematic studies in rocks showing a large variation in second-phase contents are missing so far. To close this gap, this study investigates a series of contact metamorphic calcite marbles with variable volume fractions of second-phase minerals (f_p of 0.0004 up to 0.5). The advantage of a contact metamorphic field laboratory is the chance to obtain temperature and time-resolved information on the microstructural evolution of polymineralic aggregates. This information is mandatory for the interpretation of metamorphic microstructures. With this perspective, we will discuss the variations in the effect of the second phases on the matrix grain size for different metamorphic temperatures as a function of changes in volume fraction and grain size of second phases as well as their dispersion. We will be able to define two different growth behaviors, which are either controlled by the matrix growth and local pinning of matrix grain boundaries by second phases or by complete pinning of matrix grains by second phases, where the coupled growth of matrix and second-phase grains results in larger grain sizes. As will be demonstrated, this distinction depends on variations in the physical pinning processes and is crucial for the interpretation of the evolution of metamorphic rocks.

2. Geological setting

The field area of this study is located northwest of Verona, northern Italy, close to the intersection between the two major Alpine lineaments, the Tonale and Giudicarie Faults (Fig. 1a). The samples investigated were taken from the Calcare di Angolo formation, a Middle Triassic sediment unit east of the Monte Re di Castello tonalite (Adamello intrusions). After a lagoonal and evaporitic milieu, a fast marine transgression in the Anis induced the deposition of the Calcare di Angolo in the form of dark gray micritic and massive (lower part) to thin bedded (upper part) limestones, which are intercalated with marly layers. In the non-metamorphosed limestones, crinoid columnal plates can be found. Contact metamorphism (Brack, 1983; Riklin, 1983), during the emplacement and cooling of the Monte Re di Castello tonalite transformed the original sediments into gray to white calcite marbles. The Monte Re di Castello tonalite is the oldest intrusion of the Adamello complex with an Eocene-Oligocene age of 42–40 Ma (Hansmann and Oberli, 1991) and consists dominantly of tonalitic to granodioritic rocks. Amphibole-rich gabbros and diorites occur