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# New polygonal grains formed by dissolution–redeposition in quartz mylonite

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**Abstract**—Arrays of polygonal grains are common along original grain margins, subgrain boundaries and intragranular fractures of deformed quartz porphyroclasts. We investigated these internal polygonal grains within quartz porphyroclasts of a partially recrystallized quartz mylonite deformed in the presence of fluid in a greenschist facies shear zone from Quadrilátero Ferrífero, southeastern Brazil. Optical observation showed profuse fluid inclusions and muscovite flakes inside the deformed porphyroclasts. Observation by SEM revealed the presence of crystal faces on the internal polygonal grains. Domainal *c*-axis analysis showed that the internal polygonal grains have a preferred orientation similar to the host porphyroclasts (*c*-axes at low angles to the stretching lineation), but different from the fabric of recrystallized matrix grains (type-1 cross girdle). Based on these observations, we suggest a mechanism of *in situ* solution–reprecipitation, where the new grains are nucleated in the intergranular fluid-filled pores present along the internal discontinuities of the porphyroclasts. The grains grow with their boundaries largely in contact with fluid, allowing the development of crystal faces, at least during the early growth stages. After the new grain reaches the opposite pore wall, further grain growth should occur through ordinary grain boundary migration mechanisms driven by the strain energy difference between the new grain and the host porphyroclast. We conclude that in these rocks, *in situ* solution–reprecipitation operated in conjunction with crystal–plastic processes, accounting for the nucleation of new grains during deformation/recrystallization in the presence of fluid under low grade metamorphic conditions. Copyright © 1996 Elsevier Science Ltd

## INTRODUCTION

Small, new polygonal grains in mylonites are generally thought to originate from strain-induced recrystallization mechanisms such as grain boundary migration and subgrain rotation where the new boundaries develop from pre-existing grain or subgrain boundaries (Drury & Urai 1990). Although such mechanisms are well documented in most mylonitic rocks, there are cases where the formation of new polygonal grains may result from direct precipitation from a solution (den Brok 1992). Some microstructures, however, are dubious and have given rise to contrasting interpretations. For example, arrays of polygonal grains are commonly seen to cross-cut quartz and feldspar porphyroclasts deformed under low grade conditions (den Brok 1992, fig. 2, Fitz Gerald & Stünitz 1993, fig. 4, Hippertt 1994a, fig. 6), generally without any visible offset across the arrays. These arrays have been interpreted either as recrystallized grains produced via strain-induced mechanisms (Tullis *et al.* 1973), or as a product of redeposition in extensional fractures (den Brok 1992). In order to investigate the origin of these controversial microstructures, we have performed an optical and Scanning Electron Microscopy (SEM) study of the small, new, polygonal grains occurring along grain margins, subgrain boundaries and fractures within deformed quartz porphyroclasts from a mylonitic quartzite, sheared in the presence of fluid in a low grade shear zone. Our data and observations indicate that these new grains have formed via local solution–reprecipitation in intergranular pore space, as previously proposed by Wickham (1972) and den Brok (1992). However, in this contribution, we wish to present a different model valid for deformation in the presence of fluid, where solution–reprecipitation is envisaged to be concurrent with crystal–plastic processes and strain-induced recrystallization.

## GENERAL DESCRIPTION

### Geological setting

The Moeda–Bonfim shear zone (Hippertt 1993) extends along the contact between a granite gneiss dome (Bonfim Complex) and a sequence of Proterozoic micaceous quartzites (Moeda quartzites) in the southern Quadrilátero Ferrífero region, southeastern Brazil. The shear zone affects both the granitic rocks and the quartzites. For this study, oriented samples of the sheared Moeda quartzites were taken in a 40 m-wide zone adjacent to the contact with the granite gneiss (see Hippertt 1994c for sample location). Geothermometric analysis of feldspars of the granitoid indicate that deformation occurred under greenschist facies metamorphic conditions ($T \sim 400$–$450$ °C; $P \sim 4$ kb). These metamorphic conditions are also reflected by the absence of crystal–plastic microstructures in feldspars. Deformation occurred in the presence of a water-rich fluid phase, as indicated by the syn-deformational enrichment of both lithologies in sericite. The amount of sericite (3–35%) increases consistently with deformation, and phyllonite layers (from centimetre- to a metre-scale) with large amounts of mica (60–90%) commonly found in the most highly strained domains of the shear zone. Strain determinations by the Fry (1979) method in quartz porphyroclasts indicate a maximum $\varepsilon_1 = 0.84$ and $\psi = 65°$. The reader is referred to Chauvet *et al.* (1994) and Chemale *et al.* (1994) for an extensive description on the geological setting.

### *Optical microstructures*

The sheared Moeda quartzite shows well-defined, anastomosing sericite-rich folia surrounding ellipsoidal aggregates of elongated polygonal quartz grains (30 to 120 $\mu$m) associated with small amounts (less than 15%) of relic quartz porphyroclasts. These micaceous domains define a typical '*S*–*C*' structure on a microscale. The grain size of the matrix quartz decreases with increasing deformation and sericite enrichment.

The porphyroclasts show great variation in shape and size (80 $\mu$m to 2 mm). Generally, some degree of undulatory extinction defined by large, elongate deformation bands (or elongate subgrains) is present parallel to the *c*-axis trace of the porphyroclast, indicating crystal–plastic deformation by basal $<a>$ slip. Planar arrays of equant, polygonal grains (10 to 70 $\mu$m), locally associated with small amounts of sericite, cross-cut some of the porphyroclasts without displacement across the bands. These arrays are generally oriented at low angles to the *c*-axis trace, and are normally continuous with the '*S*' planes in the matrix (Fig. 1a). Fluid infiltration along the internal subgrain boundaries and fractures is indicated by the profusion of fluid inclusions and tiny phyllosilicate flakes (sericite and chlorite) in these interfaces (Fig. 1b). New, equant polygonal grains ($<10$ $\mu$m), optically unstrained, occur both along the porphyroclast margins and internally along subgrain boundaries and fractures (Fig. 1c). The transition from deformed porphyroclast to new grains is generally abrupt. Core-and-mantle microstructures are not common in these porphyroclasts and typical 'bulges' were rarely observed along the internal grain boundaries.

### *SEM microstructures*

SEM observations (Secondary Electron Mode) were made on fresh fracture surfaces of quartzite samples, sputter coated with a 30 *n*m-thick gold film. The quartz porphyroclasts appeared in 3-D as large, irregular-shaped grains surrounded by smaller quartz grains and tiny mica flakes. Small ($<10$ $\mu$m) faceted grains commonly occur within smooth concavities on the porphyroclast surface. Figure 2(a) shows a small quartz grain with well-developed prismatic and rhombohedral faces grown in a pore on the margin of a porphyroclast. Observation of the matrix aggregates also revealed the presence of small faceted quartz grains ($<5$ $\mu$m) grown within intergranular pores and surrounded by an aureole of pore space (Fig. 2b). These small faceted grains are preferentially associated with the matrix quartz grains of the micaceous domains where an abundant presence of interfacial pores occurs. Hippertt 1994b,c gives a detailed description of these pores. Some porphyroclasts that were big enough to be separated from the rock were broken and the fragments investigated by SEM. Many of the fracture surfaces were conchoidal as typical for quartz. In some cases, however, the parting occurred along the prismatic subgrain boundaries and/or pre-existing fractures in these porphyroclasts, revealing a highly porous interface that allows a three-dimensional view of the fluid inclusion arrays such as those optically shown in Fig. 1(b). Faceted new grains were commonly found with their bases on such porous interfaces (Fig. 2c).

### *C-axis fabrics*

Most relic quartz porphyroclasts have *c*-axes oriented at low angles to the '*S*' or '*C*' surfaces, defining a diffuse girdle with maxima at low angles to the stretching lineation and/or close to the *Y*-axis of finite strain (Fig. 3a). This fabric is nearly perpendicular to the type-1 cross girdle fabric shown by recrystallized grains of the matrix (Fig. 3b). Type-1 cross girdle fabrics are known to be produced by crystal–plastic processes during non-coaxial strain histories, where basal-, prismatic- and rhomb-slip planes were activated (Lister & Hobbs 1980). This cross girdle fabric was obtained in mica-free domains of the matrix; in the micaceous domains, the increasing presence of mica is associated with progressive development of an *X*-maximum in the *c*-axis fabrics (see Hippertt 1994a,c for discussion). The relic porphyroclasts show orientations that are in fact the most unfavourable for deformation by basal $<a>$ and/or prism $<a>$ slip. They were interpreted to represent the remnants of the original quartz grain population left behind after selective recrystallization (Hippertt 1994a). The external and internal new polygonal grains associated with these porphyroclasts show a similar *c*-axis fabric (Fig. 3c), suggesting a control of the host porphyroclast on the nucleation of the new grains. This is particularly clear when the *c*-axes are plotted separately in relation to each parent porphyroclasts (Fig. 3d).

## DISCUSSION

The most common recrystallization mechanisms involve migration of pre-existing grain boundaries and/or progressive misorientation of subgrain boundaries, since the spontaneous rearrangement of a cluster of atoms in a new orientation—a process known as classical nucleation recrystallization—is extremely unlikely to occur because of the high levels of driving energy necessary for the nuclei to survive and grow (Gottstein & Mecking 1985). Dynamic recrystallization by migration of grain boundaries usually leads to the formation of bulges and, characteristically, the new grains are considerably smaller than the host grains (Poirier & Guillope

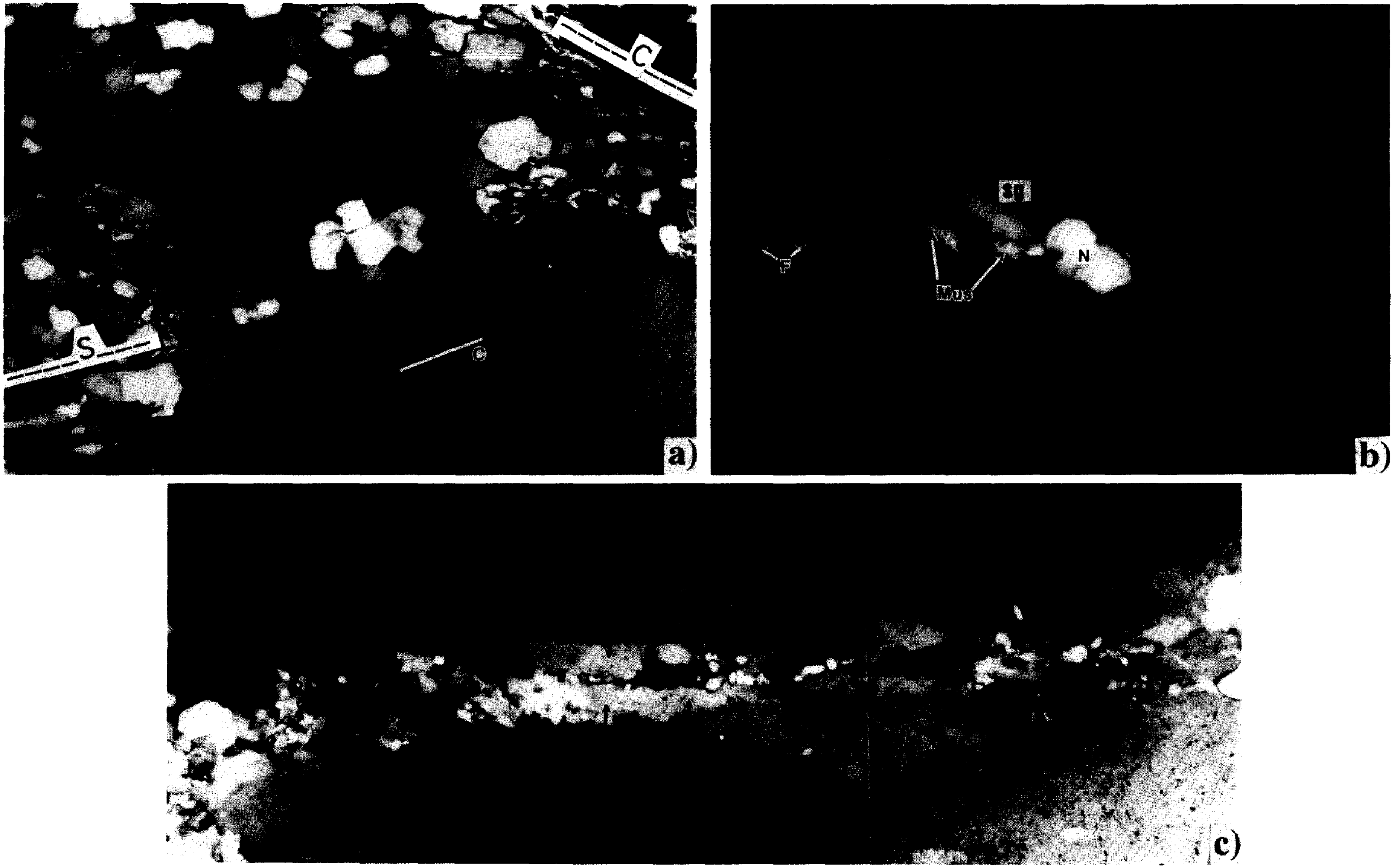

Fig. 1. Optical microstructures (crossed nicols). (a) Planar arrays of new polygonal grains cross-cutting a quartz porphyroclast along sub-prismatic planes in continuity with *S*-foliation of the matrix. Undulating bands are parallel to the projection of the *c*-axis. Such arrays of polygonal grains have been interpreted either as recrystallized grains produced via strain-induced mechanisms or as a product of precipitation in fractures. Orientations of *S–C* foliations and *c*-axis projection are indicated. Base of the photo is 0.8 mm. (b) Detail of an elongate subgrain (SG) with fluid inclusions (F) and tiny muscovite flakes (mus) along the subgrain boundaries. A small 'new' grain (N) has grown in the subgrain boundary as well. Base of the photo is 45 $\mu$m. (c) New grains occurring along a fracture in a quartz porphyroclast. Note the zone with different optical orientation adjacent to the fracture (arrows). Short side of the photo is 180 $\mu$m.

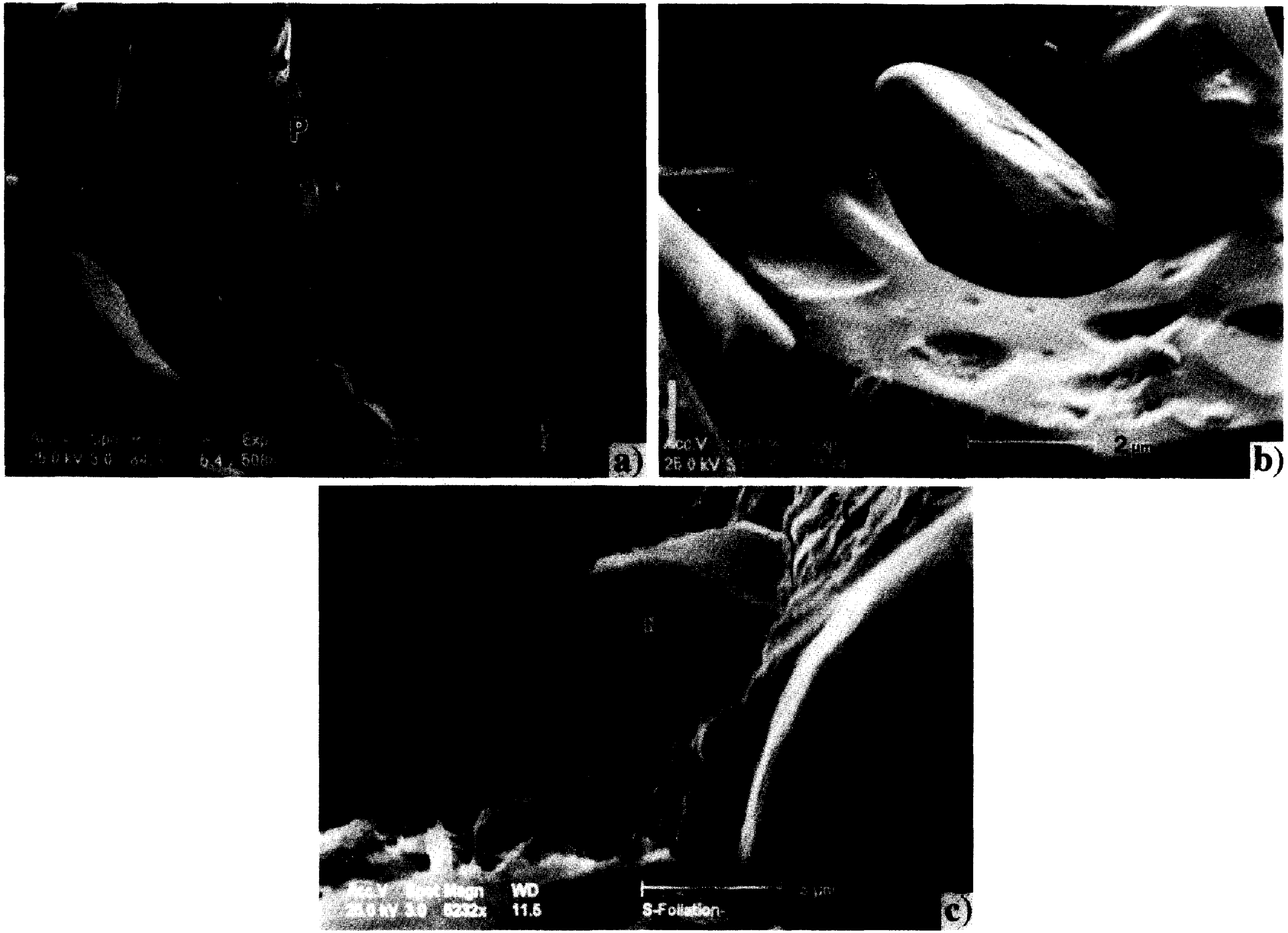

Fig. 2. SEM microstructures. (a) Small, new quartz grain with well-developed prismatic and rhombohedral crystal faces (N) adjacent to a large, deformed quartz porphyroclast (P). This new grain occurs in a concavity of the porphyroclast surface. The geometric cavities in the porphyroclast (arrow) probably correspond to new grains that were plucked off during the sample preparation. (b) Detail of a new quartz grain (n) with incipient crystal faces adjacent to an older quartz grain in which a number of individual fluid pockets are seen (f). The new grain is surrounded by an aureole pore space (arrow). Photograph taken in a quartzose domain surrounded by sericite-rich folia (se). (c) Faceted new grain (N) nucleated on a porous fracture wall (fr) within a quartz porphyroclast. This porous surface is a 3-D view of fluid inclusion arrays such as those shown in Fig. 1b.

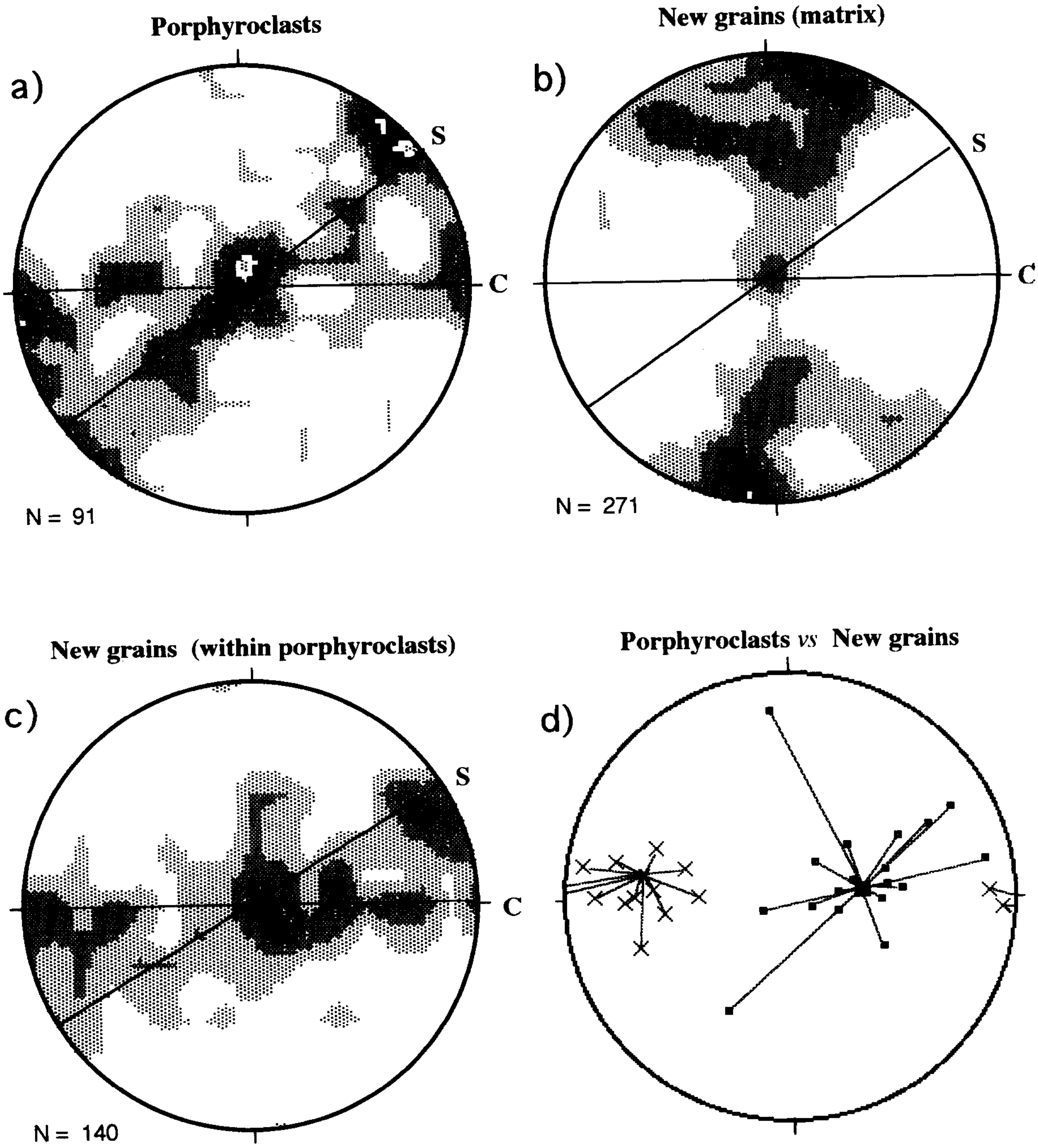

Fig. 3. Quartz *c*-axis fabrics of the Moeda quartz mylonite. (a) Porphyroclasts. (b) Polygonal new grains in the matrix (like those seen around the porphyroclast in Fig. 1a). (c) New grains occurring along the margins and inside porphyroclasts. (d) Plot of two quartz porphyroclasts (larger cross and square) with their respective internal new grains. Density contours 1, 2, 3 and 4% per 1% area in all diagrams.

1979, Urai *et al.* 1986, Hirth & Tullis 1992). Grain boundary bulging is a common recrystallization mechanism in quartz under very low grade conditions (Drury *et al.* 1985, Schmid *et al.* 1987), normally producing core and mantle microstructures (Fitz Gerald & Stünitz 1993). Although in this study the new grains within the porphyroclasts are generally smaller than the subgrains, it is unlikely that grain boundary migration has taken place since bulges, indentations (serrate grain boundaries) and typical core–mantle microstructures were only rarely observed. Furthermore, it is not clear how this mechanism could account for the formation of faceted new grains. Note also that the temperature conditions normally attributed for bulging in Quartz (sub-greenschist facies) are considerably lower than those present in the Moeda–Bonfim shear zone.

Subgrain rotation is the recrystallization mechanism typical for quartz deformed under greenschist facies conditions (White 1977). In this mechanism, the new grains are generated by progressive misorientation of subgrain boundaries resulting in a similarity of shape and dimensions between new grains and precursor subgrains (White 1979). Such a similarity, however, does not occur in the porphyroclasts investigated in this study, at least on the optical scale, where the large, elongated subgrains parallel to the *c*-axes traces clearly contrast with small, equigranular new grains, making rotation recrystallization unlikely in this case.

Fluid infiltration along internal discontinuities of porphyroclasts, promoting local dislocation glide and climb (hydrolytic weakening) and consequent recrystallization (Kronenberg *et al.* 1990, Fitz Gerald *et al.* 1991) could explain the internal arrays of new grains. Indeed, hydrolytic weakening may have occurred along some fractures in the studied porphyroclasts. This process might account, for example, for the zones of slightly different optical orientation adjacent to some fractures as shown in Fig. 1(c). However, similar to the other recrystallization mechanisms discussed above, there is no apparent reason for the development of crystal faces during recrystallization associated with hydrolytic weakening.

Direct precipitation from a fluid is a process that agrees better with our observations. If new grains nucleated on the walls of fluid-filled pores, growing with their boundaries largely in contact with fluid, they enabled the development of crystal faces during initial growth stages (Fig. 4a). Interfacial pores and intragranular fluid inclusions with dimensions in the order of a few microns are common in these rocks (Hippertt 1994b,c) and pores measuring up to 25 $\mu$m in length have been found. The new grains that show well-developed crystal faces have a grain size interval between 1 and 10 $\mu$m, consistent with nucleation and growth in these interfacial pores. New grains larger than 10 $\mu$m are not faceted, possibly because their size exceeded the pore limits. When the new grains touch the opposite pore wall, necking down of the surrounding fluid film and obliteration of the crystal faces should occur.

The most probable driving force for quartz precipitation is the supersaturation of silica in the pore fluid. Supersaturation may be instantaneously attained in the pore when communication with the matrix fluid occurs. Silica supersaturation of the matrix fluid is indicated, for example, by precipitation of quartz in the gaps formed during pull-apart of some broken quartz porphyroclasts. Evidence for access of matrix fluid into the deformed porphyroclasts is given by the common growth of mica along the subgrain boundaries. However, concurrent plastic deformation might restore the isolation of the pore, with the crystal–fluid equilibrium in the pore space being promptly re-established (Watson & Brenan 1987). Further growth of the new grain will proceed slowly in conjunction with punctual, selective dissolution along dislocation lines in the pore wall (Lasaga 1983, Brantley *et al.* 1986). Even when no communication with the matrix fluid occurred, dissolution along dislocation lines with subsequent solution-transfer across the fluid-filled pore (Hippertt 1994b) may also account for the nucleation of new grains in pores lying between old grains with

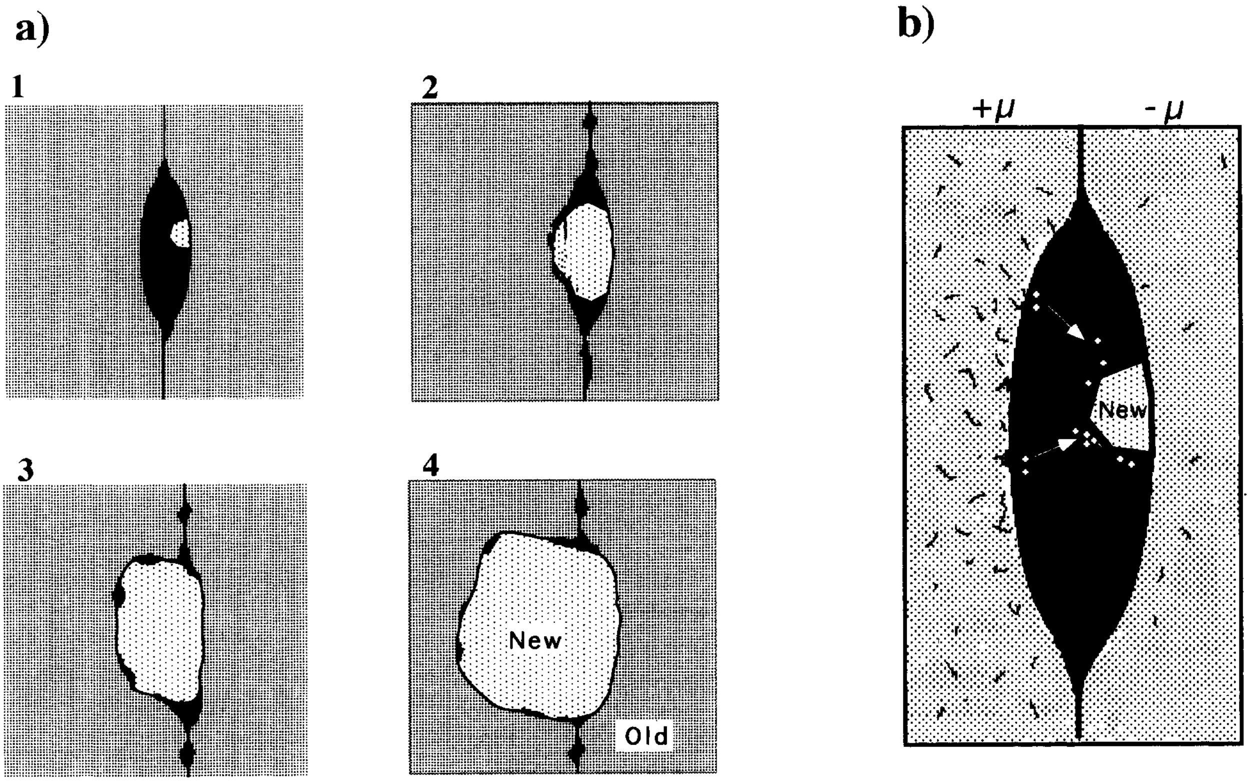

Fig. 4. (a) Suggested mechanism of local solution–redeposition. New grains are nucleated within intergranular, fluid-filled pores (stage 1) growing with their boundaries largely in contact with fluid enabling development of crystal faces, at least during the initial growth stages. After contacting the opposite pore wall (stage 2), the growing new crystal progressively necks down the surrounding fluid, increasing the area of the 'dry', solid–solid interfaces with the old grains, consequently inhibiting the mechanism of dissolution–reprecipitation (stage 3). Further growth (stage 4) occurs with increasing participation of strain-induced grain boundary migration driven by the dislocation density contrast between old and new grains. (b) Sketch showing solution transfer of silica across an isolated fluid pocket between two quartz grains with different levels of stored distortional energy. A gradient of chemical potential ($\mu$) induces dissolution of the most strained grain, diffusion across the fluid (arrows) and redeposition in a new grain nucleated on the opposite pore wall.

different dislocation densities. In this situation, a gradient of chemical potential between the two pore walls induces dissolution of the most strained grain, diffusion across the pore fluid and redeposition in the opposite pore wall (Fig. 4b). With the progressive growth of the crystal and necking down of the surrounding fluid, dissolution in the pore wall should diminish, as the boundary between the new grain and porphyroclast progressively becomes a dry, solid–solid interface. Then, the stored strain energy gradient between the new grain and the deformed host porphyroclast should become the main driving force for grain growth via grain boundary migration.

Summarising, we suggest that solution–precipitation and crystal-plasticity were concurrent processes in these rocks during low grade deformation in the presence of fluid. Crystal-plasticity produced discontinuities such as subgrain boundaries and deformation bands along which fluid-filled pores were formed. Solution–reprecipitation accounted for nucleation and growth in the pore space with subsequent grain growth by strain-induced grain boundary migration. Deformation microstructures such as subgrains and deformation bands, as well as the type-1 cross girdle fabric shown by the matrix grains are evidence for operation of crystal–plastic processes. Evidence for fluid activity includes the development of phyllonites and dissolution cleavages, the truncation of quartz porphyroclasts by dissolution seams and the presence of fibrous quartz in some cracks and gaps between separated porphyroclast fragments (Hippertt 1994a).

It should be noted that grain growth in pores should not produce preferential orientation of *c*-axes at low angles with the main growth direction, as usually occurs during crack sealing (Cox & Etheridge 1983). Instead, in a pore with a stable geometry in relation to fluid phase composition and physical conditions (i.e. where an equilibrium dihedral angle has been attained; see Watson & Brenan 1987, Lee *et al.* 1991), the new grains may be syntaxially nucleated on the pore wall, hence developing crystallographic orientations close to that of the host porphyroclast. This is indicated by similar fabric skeletons of porphyroclast relics and new grains. In fact, some of the small new grains could have the same orientation as the host and may not be visible optically, causing an underestimation of their number (den Brok, 1996, written communication). In addition, similar *c*-axis fabrics of host and new grains are also produced by subgrain rotation and grain boundary migration–recrystallization mechanisms and therefore cannot be used to infer precipitation.

## CONCLUSIONS

Although our data do not permit quantitative inferences on deformation partitioning between crystal-plasticity and solution–reprecipitation in the rock investigated, two main general conclusions can be drawn with respect to the participation of solution–transfer processes in the evolving microstructure of mylonites:

(1) Mass-conservative solution–reprecipitation at a grain scale can be an effective initiation for subsequent grain growth through grain boundary migration, leading to the formation of polygonal aggregates identical to those produced exclusively by solid state recrystallization. Solution–precipitation and crystal-plasticity may be concurrent processes that assist each other during deformation in the presence of fluids under low grade conditions.

(2) The proposed mechanism of *in situ* solution–reprecipitation offers a reasonable explanation to the intriguing bands of recrystallized grains that commonly cross-cut deformed porphyroclasts without any off set across the bands.

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