Can quartz deform by dislocation creep?
The evidence for dislocation creep in quartz reviewed.

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INTRODUCTION

When I first heard why mylonites were called mylonites (see Lapworth, 1885) I was quite astonished. What had milling to do with mylonites? I was even more astonished when I read how Lapworth (1885) described quartz deformation microstructures from the Moine Thrust mylonites. He wrote, for example, "the original crystals are crushed and spread out, and new secondary minerals, mica and quartz, are developed", or, "[the crystals] are crushed, dragged, and ground out into a finely-laminated schist (Mylonite, Gr. mylon, a mill) composed of shattered fragments of the original crystals of the rock set in a cement of secondary quartz." How could he have so completely misinterpreted the microstructures in these rocks, I wondered. Had he accidentally taken samples from a cataclastic fault zone? It took a while before I found out that in Lapworth's times, undulatory extinction, subgrains, and recrystallised grains, were considered to be cataclastic microstructures, and that this had been the standard interpretation of these microstructures from the earliest beginning of optical microscopy (e.g., Rosenbusch, 1885) until about the early nineteen sixties.

Several years later, Chris Spiers and I were involved in an experimental study to find out how water makes quartz weak and ductile at high P-T conditions.

Previous experiments on quartzite had yielded apparently contradictory results: deformation microstructures suggested that water would promote dislocation creep, whereas mechanical data suggested that water would promote diffusional creep (Jaoul et al., 1984; Kronenberg and Tullis, 1984). In our own experiments, however, quartzites appeared to be weakened and made ductile by solution-precipitation creep (SPC) and stress-corrosion microcracking (SCM) (den Brok and Spiers, 1991; den Brok, 1992). The deformation microstructures due to SPC and SCM only *looked* like dislocation creep microstructures, but were not due to dislocation creep. SCM lead to cataclastic fragmentation of the quartz. The fragments translated and rotated slightly, and then healed again. A subgrain-like structure developed in this way. Fine new grains had precipitated in open microcracks and looked like recrystallised grains. According to us these microstructures had possibly been misinterpreted as dislocation creep structures in previous studies.

I only later realised that we had unintentionally provided experimental evidence for Lapworth's (1885) point of view, i.e., for the cataclastic interpretation of what are nowadays considered crystal-plastic deformation microstructures, and that this had serious consequences. It meant that deformation by two entirely different deformation mechanisms, (i) SPC plus SCM and (ii) dislocation creep, may lead to the same kind of deformation microstructures.

How can we distinguish between the two? Can we say, for example, that if subgrains look such and such they developed by SPC and SCM and if they look so and so they developed by dislocation creep? It seems obvious that this question may be answered by comparing deformation microstructures in experiments where quartz deformed by dislocation creep with those in experiments where quartz deformed by SPC and SCM. The problem is, however, that in all of the experiments reported in the literature, the microstructures themselves were used

as an argument for dislocation creep. There are no experiments in which it was demonstrated *independent* of the deformation microstructures that dislocation creep was the dominant deformation mechanism.

How can we demonstrate in an experiment that quartz deformed by dislocation creep? To answer this question, we would ideally want to do an in-situ deformation experiment in which we would be able to see dislocations nucleate, see them glide and climb, and come together in dislocation walls. We would need to be able to count the dislocations, determine their contribution to the total strain and then compare this with macroscopically measured strain. It needs no explanantion that such an experiment cannot technically be carried out.

The next best method would be to test a microphysical dislocation creep model, but even this has not been done. It cannot be done because we do not understand *how* quartz deforms by dislocation creep (e.g., Paterson, 1989). In fact, the arguments used to demonstrate that quartz deformed by dislocation creep in experiments are indirect and inconclusive. They include: (i) the optical microstructures themselves, (ii) transmission electron microcopy (TEM) observations of dislocation microstructures, and (iii) the occurence of lattice preferred orientations (LPO's) of flattened quartz grains. None of these arguments has been demonstrated experimentally to be conclusive! The microstructures, the TEM observations and the LPO's may be *consistent* with dislocation creep, but they cannot be used as definitive evidence for its dominance or even occurrence.

This brings us to the central question of this paper: how sure can we be anyway that quartz *can* deform by dislocation creep, if the evidence for it is open to discussion? Maybe Lapworth (1885) was right and we are wrong today. I will show below that we cannot be sure about the answer. This means that the "old" idea of ductile deformation by cataclastic deformation in the sense of Lapworth (1885) should be re-considered as a serious alternative mechanism to explain the

occurrence of "crystal-plastic" microstructures in deformed quartz rocks. Dislocation creep may, consequently, be a much less important deformation mechanism in quartz rocks in the nature than is commonly thought.

EXPERIMENTAL WORK ON QUARTZ

One of the arguments that quartz can deform by dislocation creep is that this has been experimentally demonstrated in experiments. In the next sections I will explain why I think that this is not the case.

The hydrolytic weakening problem

It is a well established fact that small amounts of water make quartz weak and ductile at high P-T conditions (e.g., Jaoul et al., 1984). This weakening effect of water was first observed by Griggs and Blacic (1964) and was (and still is) firmly believed to be an intracrystalline effect (e.g., Paterson, 1989). The idea is that the water makes quartz deformable by dislocation creep. The problem is that we do not understand *how* water makes this possible. This is a problem for almost 35 years now. Water must (i) diffuse into the quartz, (ii) promote nucleation of dislocations, (iii) promote glide of dislocations, and (iv) promote climb of dislocations, and we do not know at all how water does all this. It is generally *believed* that it has to do something with the hydrolysis of relatively strong Si-O-Si bonds to form weaker Si-OH:HO-Si bonds, whence *hydrolytic* weakening (see Paterson, 1989, for a review of the problem), but there is no conclusive evidence for this. In my opinion the hydrolytic weakening problem may possibly be solved

if it is assumed that water promotes SPC and SCM instead of dislocation creep (den Brok and Spiers, 1991).

Dry quartz single crystals

"Dry" quartz is extremely strong and brittle (Blacic and Christie, 1984). At room P-T conditions it can easily sustain ~2.4 GPa differential stress without measurably deforming. Only at very high pressures and temperatures (typically of the order of 1.5 GPa and 800°C) dry quartz may be deformed "plastically", at very high differential stress (typically 1.2 GPa) and to a few percent strain only (e.g., Blacic and Christie, 1984). Under these conditions quartz deforms dominantly by slip along deformation lamellae oriented sub-parallel to planes of high shear stress. How the lamellae form and how this slip takes place is not well understood. TEM-observations show that lamellae that develop under such conditions consist of ~50 nm wide zones of glass and are locally associated with "festoons" of dislocations, suggestive of defectless yielding (Christie and Ardell, 1974).

Wet quartz single crystals

In the presence of small amounts of added water (typically 0.1 to 1 vol%) and at high P-T conditions (such as above) quartz single crystals may easily be up to 2 orders of magnitude weaker than dry crystals at otherwise similar experimental conditions (e.g., Ord and Hobbs, 1986). First it was thought that the water would diffuse into the quartz and cause hydrolytic weakening (Griggs and Blacic, 1964), but it was found out later that (i) the crystals are weakened essentially by SCM (Kirby and Kronenberg, 1984; Kronenberg et al., 1986; FitzGerald et al., 1991) and (ii) deformation takes place without measurable

uptake of water in the crystal lattice (Kronenberg et al., 1986; Gerretsen et al., 1989).

FitzGerald et al. (1991) studied the microstructure of such crystals in great detail. They found that strain was almost entirely confined to the ends of the crystals, where complete recrystallisation into a fine-grained aggregate had taken place. The rest of the crystal contained abundant microfractures. Dislocations developed only in the vicinity of these fractures (and maybe in the fine-grained aggregates). FitzGerald et al. (1991) suggested that the dislocations developed during the fracturing or healing process. The dislocation structures showed "local transitions from regions of easy glide and recovery into regions where no dislocations were observed". These were therefore assumed to have contributed to minor (<0.5%) local crystal plastic strain next to the fractures. The observations show that dislocations can be formed during deformation of initially dislocation-free quartz, but they can hardly be taken as evidence for dislocation *creep*. The dominant weakening effect in the experiments was either due to SCM, or due due to deformation processes taking place in the fine grained aggregate at the sample ends.

Fine-grained quartz aggregates

When fine-grained quartz aggregates (flint or novaculite) are deformed dry, these are almost as strong and brittle as dry single crystals. When they are wet they can be deformed ductile at low stresses (Green et al., 1970; Kronenberg and Tullis, 1984; Mackwell et al., 1989). The deformation mechanism active during the wet deformation is improperly understood. The grains are too fine to see what happened with them, and samples completely recrystallise during deformation. Deformation *may* have occurred by dislocation creep (Kronenberg and Tullis,

1984). TEM observations are consistent with this idea, but the strain associated with the observed dislocation structures may be very small and most of the deformation *may* alternatively have taken place by other mechanisms. According to Mackwell et al. (1989) deformation occurs by SPC because the mechanical behaviour is consistent with this mechanism.

In these fine-grained aggregates, relatively strong LPO's are commonly developed during deformation. This may be suggestive of dislocation creep (e.g., Kronenberg and Tullis, 1984), but according to Green et al. (1970) these LPO's are the result of strain-induced recrystallisation at low temperature, and stress controlled selective normal grain growth at higher temperature. The latter is consistent with deformation by SPC.

Coarse-grained quartz aggregates

Dry coarse-grained quartz aggregates (quartzites) also behave like dry quartz single crystals and fine-grained aggregates and are extremely strong and brittle. With increasing amounts of added water they become increasingly weak and ductile (e.g., Jaoul et al., 1984; den Brok and Spiers, 1991). Not only strength, but also the stress exponent *n* for power law creep decreases with increasing water content, from *n*-values of more than ~10 at very low water content, to *n*-values as low ~1.3 at relatively high water content (e.g., ~1 vol%; Jaoul et al., 1984; den Brok and Spiers, 1991). This suggests that water promotes a diffusional deformation process such as SPC. A similar water-weakening effect associated with decreasing stress exponents due to SPC was observed in sandstone by Rutter and Mainprice (1978) and in rocksalt by Urai et al. (1986).

With increasing water content, the deformation microstructures are characterised by a decreasing significance of deformation lamellae, and an

increasing significance of "subgrains" and of small new "recrystallised" grains. According to Tullis et al. (1973) and Jaoul et al. (1984) this is due to strain-induced recrystallisation and/or rotation recrystallisation, which means that water must have diffused into the grains and promoted nucleation, glide *and* climb of dislocations. According to den Brok and Spiers (1991) these microstructural changes are due to increased activity of SCM and SPC, the latter leading to precipitation of new grains in microfractures oriented parallel to the axial shortening direction (*Z*). The dislocation substructures, however, are consistent with dislocation creep (e.g. Tullis and Yund, 1989; den Brok 1992). It remains the question, though, how much crystal plastic deformation is associated with these structures. Fourier transform infra-red (FTIR) spectroscopy measurements of intracrystalline water content of the deformed grains before and after deformation show that water weakening occurs without a measurable uptake of water by the grains (Kronenberg and Wolf, 1990; den Brok et al., 1994). This is consistent with weakening by SPC and SCM.

Perhaps the strongest argument that dislocation creep occurs in experimentally deformed wet quartzites is the presence of LPO's of c-axes of *flattened original grains*. The presence of such LPO's has been reported, a.o., by Tullis et al. (1973), Jaoul et al. (1984), and Dell'Angelo and Tullis (1986). Though, care should be taken. These LPO's have an *orthorhombic* symmetry, with point maxima at about 30-45° to *Z* and lying in the plane of the thin section at the rim of the pole figure. As pointed out by Dell'Angelo and Tullis (1986) one would expect the LPO's to exhibit *axial* symmetry, because the c-axes distribution was random before deformation and samples were axially flattened. Dell'Angelo and Tullis (1986) showed, that this othorhombic symmetry was due to an error in the measurement technique. C-axes were measured on a universal stage without having taken care that *all* the grains within a given area were measured. Starkey

and Sutherland (1978) have shown that in case of a random c-axes distribution, orthorhombic LPO's may be obtained due to selective sampling of grains that show the highest birefringence. This means that the LPO's are probably much weaker. LPO's of Den Brok and Spiers (1991) also showed this orthorhombic pattern. Den Brok (1992) re-measured these samples, now carefully measuring *all* the grains within a given area, and then there appeared to be no siginifant LPO anymore. So, it remains to be demonstrated that old-grain LPO's *do* form.

Water containing synthetic quartz

Most deformation experiments aimed at understanding of hydrolytic weakening have been carried out on water-containing synthetic single crystals. This has the (apparent) advantage that water does not have to diffuse into the crystals. McLaren et al. (1989) have demonstrated, however, that this material is not a good analog for real quartz. Water in synthetic quartz is present under high pressure in nanometer-scale ellipsoidal inclusions oriented with their long axes parallel to the prismatic planes. Even without deformation, but on heating, these inclusions decripitate by development of rhombic microcracks and send out prismatic dislocation loops with orientations related to the shape of the inclusions. Such high-pressure ellipsoidal inclusions are conspicuously absent in natural quartz rocks (Kronenberg et al., 1990).

It is interesting to note that these crystals even *when* they are full of dislocations after heating can be deformed plastically only to very low strains before fracturing occurs (typically <5%; e.g., Linker et al., 1984).

Conclusion concerning experimental work

The experimental work on quartz shows that water makes quartz weak and ductile (at appropriate P-T conditions). It also shows that water promotes stress corrosion microcracking (SCM), recrystallisation, and the development of a dislocation microstructure that is consistent with dislocation creep. Mechanical data suggest that water promotes a diffusional deformation mechanism such as solution-precipitation creep (SPC), and FTIR measurements of intracrystalline water content before and after deformation show that water weakening occurs without the uptake of measurable amounts of water. Strong LPO's develop in fine-grained aggregates where recrystallisation is important. Weak (or no) LPO's develop in coarse grain aggregates.

It may be clear from this that we are still far from understanding what really happens in these experiments. Many observations may be consistent with dislocation creep, but the experiments can certainly not be used as evidence that quartz *can* deform by dislocation creep. How about the other arguments for dislocation creep?

OPTICAL MICROSTRUCTURES

Can we say: "the quartz grains show undulatory extinction, subgrains, recrystallised grains, so these grains deformed essentially by dislocation creep"? No, I do not think so. It remains unclear in many cases whether or not these structures may have developed by fracturing, slight rotation and translation of the fragments and subsequent healing such as thought, e.g., by Lapworth (1885). The fractures may have changed position slightly by disssolution and precipitation. Small new grains may have grown in fractures, or alternatively, by stress-induced recrystallisation, or strain-induced recrystallisation, the latter possibly driven by

differences in density of dislocations developed during the fracture process. Fluid inclusions at the fracture boundaries may have disappeared. How could we tell the difference between these and "real" dislocation creep microstructures? There are no experiments in which it is demonstrated that these structures develop by dislocation creep. It must be concluded that these microstructures are *consistent* with dislocation creep but cannot be used as conclusive evidence for it.

TRANSMISSION ELECTRON MICROSCOPY

The observation of TEM microstructures like free dislocations ans subgrain walls is certainly consistent with dislocation creep, but is their presence conclusive proof? We do not know enough about these structures to be really sure that repeated nucleation, glide and climb of these dislocations lead to crystal plastic strain. The dislocations may alternatively be related to fracturing and healing processes, or to crystal growth under stress. These phenomena have not been studied under such high P-T conditions. Moreover, the crystal plastic strain associated with these structures may be very small and deformation may have been accomodated largely by other mechanisms. We need to further systematically study the development of dislocation structures in wet quartz under high P-T conditions.

LATTICE PREFERRED ORIENTATIONS

Can LPO's be used as an argument for dislocation creep? Certainly, at present, it may be the most likely explanation, in many cases, but it is not

conclusive evidence. There are other mechanisms that may lead to LPO's, such as, for example, stress-induced recrystallisation or grain growth (Hobbs, 1968; Green et al., 1970; Shelley, 1989), crystal growth, e.g. in slickensides or fibrous veins (Cox and Etheridge 1983), or even pressure solution (Hippertt 1994).

Lister et al. (1978) succesfully modelled at least *some* common LPO's, assuming that grains deformed by glide of dislocations. These LPO's may provide the strongest argument for dislocation creep. Though, again, care should be taken. First, these models are not supported by experiments. Second, Lister et al. (1978) have used information about active slip systems that were based on (i) the orientation of deformation lamellae (Christie and Green, 1964) and (ii) TEM observations of slip systems in experimentally deformed *synthetic* crystals. Both of these information sources are questionable. It is well known now, that there is no simple relationship between the orientation of the lamellae and the orientation of the dislocations found within them (White, 1973). As for the synthetic quartz, it may very well be that the lensoid shape of the high pressure fluid clusters oriented parallel to the prismatic plane and subsequent decrepitation have determined the orientation of the dislocations that developed in these samples (cf. McLaren et al., 1989). We do not know.

CONCLUSION

The experimental work on quartz does not provide unequivocal, conclusive evidence that quartz can deform by dislocation creep. Moreover, the commonly used observations as argument for dislocation creep, including (i) optical microstructures like undulatory extinction, subgrains and recrystallised grains, (ii) TEM observations of dislocation substructures, and (iii) Lattice preferred

orientations, may be consistent with dislocation creep, but are not conclusive, unambiguous arguments.

Experimental work has shown that quartz can only be deformed ductile to a significant degree when small amounts of water are present, but the water makes quartz ductile is not understood. It may make dislocation creep possible *or* stress corrosion microcracking and solution-precipitation creep. Maybe water promotes both of these processes. We do not know. We cannot be sure whether quartz can deform by dislocation creep or not. We should be carefull with the interpretation of 'crystal plastic' deformation microstructures in quartz. The 'old' idea of ductile deformation by cataclastic deformation in the sense of Lapworth (1885) and associated with solution transfer processes should therefore be re-considered as an alternative mechanism to explain these microstructures.

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