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Abstract

Numerous rod-shaped calcite crystals occur in the blueschist to eclogite facies marbles of Syros, Greece. The rods show a shape-preferred orientation, and the long axes of the rods are oriented at a large angle to foliation. The crystals also have a crystallographic-preferred orientation: calcite c-axes are oriented parallel to the long axes of the rods. Based on their chemical composition, shape, and occurrence in high-pressure marbles, these calcite crystals are interpreted as topotactic pseudomorphs after aragonite that developed a crystallographic-preferred orientation during peak metamorphism. This interpretation is consistent with deformation of aragonite by dislocation creep, which has been observed in laboratory experiments but has not been previously reported on the basis of field evidence. Subsequent to the high-pressure deformation of the aragonite marbles, the aragonite recrystallized statically into coarse rod-shaped crystals, maintaining the crystallographic orientation developed during deformation. During later exhumation, aragonite reverted to calcite, and the marbles experienced little further deformation, at least in the pseudomorph-rich layers. Some shearing of pseudomorph-bearing marble layers did occur and is indicated by twinning of calcite and by a variable inclination of the pseudomorphs relative to foliation.

KEYWORDS: aragonite, pseudomorph, Greek Aegean Islands, high-pressure metamorphism, marble, deformation, dislocation creep

1. Introduction

Aragonite, a high-pressure polymorph of calcite, is the stable calcium carbonate mineral during metamorphism at blueschist to eclogite facies conditions (Carlson, 1983). Nevertheless, aragonite is rare in high-pressure marbles because it transforms to calcite during exhumation, unless the temperature is unusually low (Carlson and Rosenfeld, 1981). Although marbles on the island of Syros (Cyclades, Greece) contain high-pressure mineral assemblages (Schumacher et al., 2000) consistent with the blueschist to eclogite facies metamorphism recorded by associated mafic rocks (Ridley, 1984; Okrusch and Bröcker, 1990), no aragonite has been identified there. Because these rocks show evidence of an incomplete greenschist facies overprint (e.g. Altherr et al., 1979), the absence of aragonite is not surprising. However, the ubiquitous presence of oriented, acicular calcite textures is both surprising and interesting. We interpret these textures as calcite pseudomorphs after aragonite for two reasons. First, the acicular shape of calcite rods is similar to the habit of aragonite. Second, these marbles experienced peak deformation at 450-500°C and 12-20 kb (Dixon, 1976; Okrusch and Bröcker, 1990; Dixon and Ridley, 1987; Bröcker and Enders, 1999), conditions well within the stability field of aragonite (Carlson, 1983).

In terms of its tectonic and geologic setting, Syros is similar to other islands in the Cyclades of Greece. Blueschist and eclogite facies mineral assemblages and fabrics in marbles and mafic meta-igneous rocks are variably overprinted by greenschist facies retrogression. Most workers regard the earlier high-pressure phase of deformation and metamorphism as a subduction-related event, and the later greenschist event as a product of exhumation by extension. The age of the high-pressure event is believed to be Eocene (40 to 42 Ma) based on Rb-Sr and K-Ar data (Altherr et al., 1979), but recent data (Bröcker and Enders, 1999) suggest ages of ~80 Ma for this event. Younger Rb-Sr and K-Ar ages ranging between 33 and 39 Ma are interpreted as partial resetting during the greenschist event (Altherr et al., 1979). We believe that aragonite replaced calcite in carbonate units during the early high-pressure event and reverted to calcite during later exhumation.

In other metamorphic terrains where aragonite crystals are partially replaced by calcite (e.g. Theye and Seidel, 1993) or calcite pseudomorphs retain aragonite crystal form (e.g. Wang and Liou, 1991), most workers have observed aragonite rods oriented sub-parallel to the foliation (Coleman and Lee, 1962; Stöckhert et al., 1999). On Syros, however, rods in calcite pseudomorphs after aragonite are consistently oriented sub-perpendicular to the high-pressure foliation. This unusual geometry raises two questions: Why are the aragonite pseudomorphs on Syros in a structural orientation different from that of aragonite described for other localities, and what do the current orientations of pseudomorphs say about the deformation during peak metamorphism and the later exhumation of the Syros marbles?

2. Aragonite pseudomorph (AP) features

We have observed aragonite pseudomorphs (AP's) in marbles across Syros (Fig. 1), although many marble layers lack them, and no clear regional distribution exists. Instead, AP distribution relates in part to lithology: easily visible AP's occur most commonly in the purest marble units. Where present, countless acicular AP's, oriented sub-perpendicular to foliation, define the texture of these marble layers as far as they can be traced. In units of pure marble that are tens of meters thick, the acicular texture is commonly penetrative at both the hand sample and outcrop scale. The abundance and organization of the AP's is as impressive as that of muscovite in a mica-rich schist (Fig. 2).

The AP lineation at most localities is sub-perpendicular to the foliation defined by compositional banding, phengite crystals, and the axial planes of isoclinal folds (Fig. 3). Across the island, the dip of foliation is generally shallow, and the plunge of AP lineation is steep. This foliation is clearly associated with the high-pressure deformation event: at some locations, omphacite grains deflect foliation, and at other locations, foliation is parallel to a glaucophane lineation. Where the AP lineation occurs adjacent to impure marbles, the lineation defined by other high-pressure minerals is at a large angle to the AP lineation. For example, Figure 4 shows the orientation of a shallow foliation with a sub-parallel glaucophane lineation in an impure marble layer. Isoclinal fold axes are parallel to the glaucophane lineation, but the AP lineation in an adjacent clean marble layer is oriented at a large angle to the glaucophane lineation. In layers that show both AP lineation and isoclinal folds, the AP lineation is perpendicular to the fold axial plane, rather than wrapping around the fold hinge (Fig. 3). More direct measurements of the angle between AP lineation and associated foliation confirm that they commonly occur at a large angle to each other; a histogram of these measurements (Fig. 5) shows a considerable range of angles. One of the smallest angles (22°) was observed in a rock containing closely spaced shear zones of fine-grained calcite (Fig. 6). In this and some other samples, the AP's have a sigmoidal shape related to deformation by shearing. Summarizing, the long axis of the AP's is oriented sub-parallel to the direction of maximum shortening as defined by foliation and at a large angle to the maximum elongation as defined by other prismatic minerals.

At the hand sample scale, individual pseudomorphs range in length from a few mm to 10 cm, with aspect ratios ranging from 3:1 to >20:1. Individual calcite crystals are rarely longer than 1 cm, so that in many cases it appears that a single aragonite crystal has been replaced by several calcite crystals. Because the pseudomorphs occur in bundles and because each pseudomorph may consist of several calcite crystals, the actual size of the original aragonite crystals is not certain. Although the AP's have the high aspect ratio of aragonite crystals, their shapes in cross section are neither convincingly orthorhombic nor pseudo-hexagonal. At the microscopic scale, the calcite crystals that constitute the AP's show relatively high aspect ratios, elongation parallel to the lengths of the AP's, and twinning (Fig. 7). Minor phases in the rod bundles are equant grains of quartz and dolomite.

The calcite crystals forming the AP’s show a moderate to strong crystallographic preferred orientation (CPO). In thin sections cut perpendicular to the AP lineation and viewed conoscopically, most crystals have a nearly-centered optic axis. For measurements on four of these thin sections using a view that represents 50° (28% of the possible orientations of the optic axis), an average of 54% (N = 368) of optic axes were in view. Of these measured grains, 75% were weakly biaxial, and the remaining 25% were uniaxial. In addition, we examined a marble layer without visible AP’s and found that it nevertheless has a CPO with calcite c-axes at a large angle to foliation. Our attempts to identify surviving aragonite met no success. Using both Feigl’s solution and Meigen’s solution (Lewis and McConchie, 1994) to test for aragonite, we observed no stain on any of the Syros samples showing AP's, although samples of aragonite from other localities did take the stain. No aragonite peaks were observed on powder x-ray diffraction patterns of AP samples.

3. Comparison with published experiments

Experiments on the transformation of aragonite to calcite due to a decrease in pressure offer constraints on the significance of the pseudomorph textures observed in the Syros marbles. Although the results of experimental studies on this transformation during deformation (Gillet et al., 1987; Snow and Yund, 1987) are difficult to reconcile, experimental studies on the transformation during static conditions have emphasized the importance of topotaxy (which preserves crystal lattice orientations) during the phase transformation. Both Brown et al. (1962) and Carlson and Rosenfeld (1981) observed that a major fraction of the new calcite crystals had c-axes parallel to the original aragonite c-axes (the c-axis of aragonite is parallel to the long axis of acicular crystals). Other experiments by Boettcher and Wyllie (1967) showed that calcite that has replaced aragonite by topotaxy is commonly biaxial with a low 2V. The biaxial character of most calcite grains strengthens our interpretation that the acicular calcite textures we document here represent AP's. Furthermore, the acicular habit and the CPO of the calcite are significant because they imply that the AP’s are topotactic calcite replacements of elongated aragonite crystals that had a strong CPO, with aragonite c-axes oriented sub-perpendicular to foliation.

Two published sets of experiments on coaxial deformation of aragonite offer guidance on understanding the origin of the inferred aragonite textures. Hacker and Kirby (1993) deformed Carrara marble in the aragonite stability field and found that aragonite crystals grew and "penetrated into calcite crystals ~ 2 - 5 μm farther" in the direction of maximum shortening, s_3 (in co-axial deformation, s_3 is parallel the direction of maximum compression, σ_1). In axial compression experiments on synthetic aragonite marble, Rybacki et al. (in press) found a c-axis CPO of aragonite also parallel to s_3. They interpreted their experiments as reflecting dislocation creep of the aragonite with (001) as the predominant glide plane, which they argued is consistent with the (001)[010] glide system for aragonite identified by Renner and Rummel (1996) in room temperature experiments. In addition, Rybacki et al. (in press) found that aragonite grains developed a shape-preferred orientation during deformation, with the long axes of grains perpendicular to both the shortening direction and the c-axis CPO. Note that the results of the two sets of experiments differ. The Hacker and Kirby experiments (1993) document aragonite shape-preferred orientation due to growth kinetics, and the Rybacki et al. experiments (in press) document aragonite CPO and shape-preferred orientation due to deformation by dislocation creep.

4. Origin of AP shape- and crystallographic-preferred orientation

Do the oriented, acicular AP's on Syros result simply from the kinetics of aragonite growth or do they represent a fabric developed during prolonged deformation of aragonite by dislocation creep? The hypothesis that the AP alignment is a growth texture is appealing for two reasons. First, the AP's are reminiscent of the fibrous growth textures commonly observed in calcite veins (Durney and Ramsay, 1973), although the AP's show a coarser habit and are commonly penetrative at the outcrop scale, rather than occurring in unique, restricted bands. Nucleation of aragonite at the boundaries of pure layers and more rapid growth parallel to c-axes, perpendicular to the layer boundaries and foliation, is certainly plausible. Secondly, this hypothesis explains most of the features of the AP's, including CPO, acicular habit, and the parallelism between calcite c-axes and the long axes of rods. A simple driving mechanism for aragonite crystal growth by this model would be the transformation from calcite to aragonite as these rocks were carried down a subduction zone. Unfortunately, this scenario is impossible to reconcile with the high-pressure deformation history of the rocks. In order to preserve the features of the AP's, the transformation/growth hypothesis would imply that very little deformation of the AP layers had occurred since the aragonite crystals first appeared, whereas there is good textural evidence for deformation in the aragonite stability field.

Any successful model for AP alignment must take into account the high-pressure deformation history of the rocks. The strong glaucophane lineation parallel to foliation and the deflection of foliation by omphacite grains signifies that the marbles deformed while at high pressures when aragonite (rather than calcite) was present. Mesoscale folds are commonly isoclinal with their axial planes parallel to regional foliation, and their axes parallel to the regional glaucophane lineation (Fig. 4). This geometric relation between folds and mineral lineation probably reflects a significant non-coaxial strain, an interpretation consistent with the analysis of the same fabrics by Ridley (1982), who understood the shallow foliation and lineation as the record of a ductile thrust system. In contrast, a recent study by Rosenbaum et al. (2002) proposed that the high-pressure deformation event on Syros was characterized by significant coaxial strain during vertical thinning. Although the kinematics recorded by the high-pressure fabrics are ambiguous, both authors agree that the fabrics are the product of high strain, consistent with our observations of these rocks.

Strong evidence for high strain in the marbles of Syros at high-pressure conditions suggests that some of the features of AP's developed because of deformation, rather than despite it. We prefer the hypothesis that the AP's represent a fabric developed as a result of prolonged deformation of aragonite by dislocation creep. This deformation mechanism can explain the strong aragonite CPO, and it is consistent with the results of experiments on the development of CPO in aragonite by dislocation creep (Rybacki et al., in press). Because these experiments have been restricted to coaxial kinematics, they do not offer a complete guide to interpreting the kinematic event recorded by AP's. The results of the Rybacki et al. (in press) experiments, however, are similar to those for calcite obtained by Wenk et al. (1987). For calcite deformed at conditions at which crystal plastic deformation is the predominant deformation mechanism, coaxially deformed samples show orthorhombic pole figures with calcite c-axes oriented parallel to the shortening axis, and non-coaxially deformed samples show monoclinic or triclinic pole figures (Wenk et al., 1987). Because the calcite and aragonite systems appear to behave similarly during coaxial deformation, they may also be similar during non-coaxial deformation. The orthorhombic relationship between foliation and the inferred CPO of aragonite in the Syros marbles may be either the product of a coaxial deformation or a non-coaxial deformation that was sufficient to produce near-orthorhombic symmetry between aragonite CPO and foliation. Therefore, the CPO of AP's does little to resolve the debate on the kinematics of high-pressure deformation on Syros. But in either case, good agreement between the large strains accumulated during the high-pressure deformation history of the marbles and the inferred CPO of aragonite make this alternate hypothesis a compelling one.

One observation that appears inconsistent with this hypothesis is the parallelism between inferred aragonite c-axis CPO and the long axes of aragonite rods. During deformation by dislocation creep, grain shapes generally record finite strain and experience elongation sub-parallel to foliation. In these marbles, however, the long axes of AP grains are oriented at a large angle to foliation. Post-kinematic coarsening (static recrystallization) of aragonite crystals may offer a resolution to this problem. If aragonite developed a strong CPO during deformation by dislocation creep, then one would expect aragonite grains to have been relatively small and equant, or slightly elongate parallel to foliation. Following deformation, a static recovery process may have allowed grains to coarsen, forming the acicular habit typical of aragonite due to more rapid growth parallel to its c-axis. According to this scenario, the strong aragonite CPO inherited from the deformation event would have controlled the orientation of the coarsening grains by providing numerous aragonite crystals with their c-axes aligned perpendicular to foliation. Such coarsening would have occurred after the high-pressure deformation event, but before the topotactic replacement of aragonite by calcite.

Finally, three features of the AP's appear to relate to deformation during later greenschist and lower grade deformation, after calcite had replaced aragonite. First, the twinning that is common in the calcite crystals that constitute the AP's (Fig. 7) probably occurred after the reversion of aragonite to calcite because twins in aragonite have a crystallographic orientation that would not match that of twins in calcite overgrowths. The second feature is the local divergence of the long axes of the AP's from an orientation orthogonal to foliation (Fig. 5). The close association between sigmoidal AP's and brittle shear zones in the marbles suggests that the variation in the orientation of the AP's is the result of non-coaxial deformation of calcite at relatively low metamorphic grade. If so, the sense of reorientation of AP's may provide a useful indicator of the sense-of-shear associated with the late exhumation event. It may also provide the opportunity to make minimum finite strain calculations for this event. Third, the existence of obvious AP's in some marble layers but not in others is intriguing. We believe it is likely that some layers accommodated significant deformation during exhumation, and that the AP textures were destroyed by deformation in these layers. It is also possible that aragonite did not coarsen in some layers, perhaps due to the presence of more silicate minerals, so that there were no acicular crystals to pseudomorph. In at least one marble layer without rod-shaped crystals, the calcite nevertheless has a CPO consistent with an original aragonite CPO orthogonal to foliation.

5. Conclusions

Some marble layers on Syros show a pronounced acicular texture in calcite. Long axes of calcite rods are regularly oriented sub-perpendicular to a foliation clearly developed during a high-pressure deformation/metamorphic event. In addition to this pronounced texture, calcite in these rocks shows a moderately strong crystallographic preferred orientation (CPO) (with c-axes preferentially oriented sub-parallel to the long axes of rods) and weak biaxiality. Based on comparison with experimental work on the stability and deformation of aragonite and calcite, we argue that both the biaxiality and acicular texture in calcite result from replacement of aragonite that itself had both a strong, acicular texture and a CPO. The penetrative CPO in these rocks appears to provide field evidence for widespread deformation of aragonite by dislocation creep, which has been explored in laboratory experiments (Snow and Yund, 1987; Hacker and Kirby, 1993; Rybacki et al., in press). Although the aragonite texture could be the result of nucleation on layer boundaries and more rapid growth parallel to the aragonite c-axis, it is unlikely that such a texture would survive the high-pressure deformation evident in these rocks. If aragonite CPO developed by dislocation creep, the acicular habit of the grains probably developed by a static recrystallization process that followed the high-pressure deformation event but preceded the reversion of aragonite to calcite. Preservation of the AP texture limits the amount of deformation experienced by these rocks during exhumation from depths where aragonite is stable.

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FIGURE CAPTIONS

Figure 1. Aragonite pseudomorph (AP) locations studied by the authors are shown on an outline map of Syros relative to the distribution of marble units. Marble boundaries are taken from the geologic maps of Hecht (1984) and Höpfer and Schumacher (1997).

Figure 2. Hand sample of marble from SE Syros (Katergaki) showing compositional banding and calcite pseudomorphs after aragonite. Foliation defined by phengite in adjacent layers is parallel to the compositional banding. Note the large angle between the compositional banding and the AP lineation.

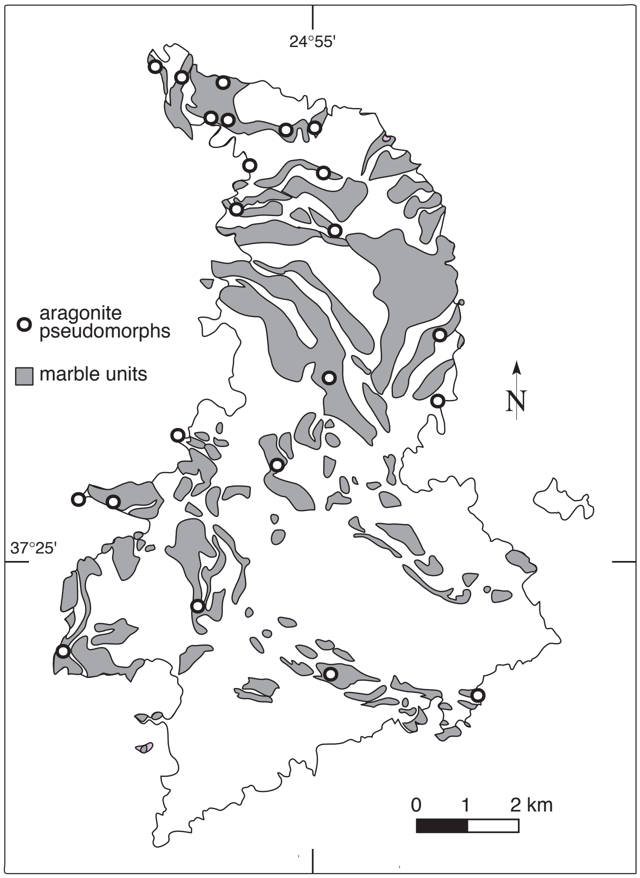
Figure 3. Isoclinal fold in marble from NW Syros showing AP lineation at a large angle to the axial plane of the fold. Glaucophane lineation in the impure marble layers is parallel to the fold axes.

Figure 4. Equal area stereographic projection of AP lineations (✶), glaucophane lineations (■), fold axes (●), and phengite foliation planes (great circles) in adjacent layers from a single location in northern Syros.

Figure 5. Histogram of measured angles between AP lineation and foliation in marbles from Syros.

Figure 6. Photomicrograph in crossed polarized light of a marble sample containing AP-bearing layers separated by fine-grained shear bands. The AP lineation is oriented at a low angle (22°) to the sheared layers.

Figure 7. Photomicrographs in crossed polarized light of an AP sample cut parallel to the AP lineation.



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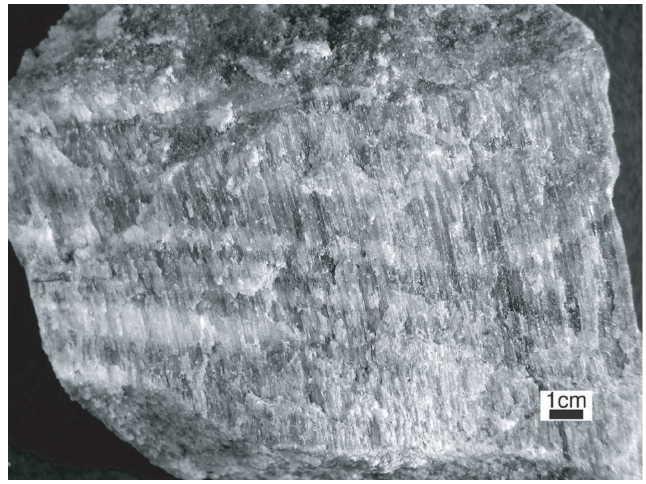

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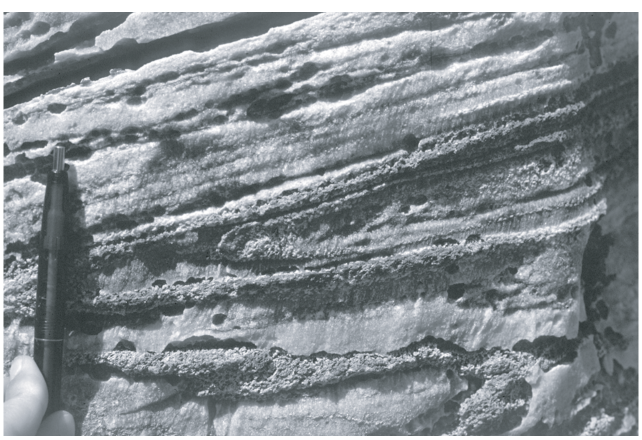

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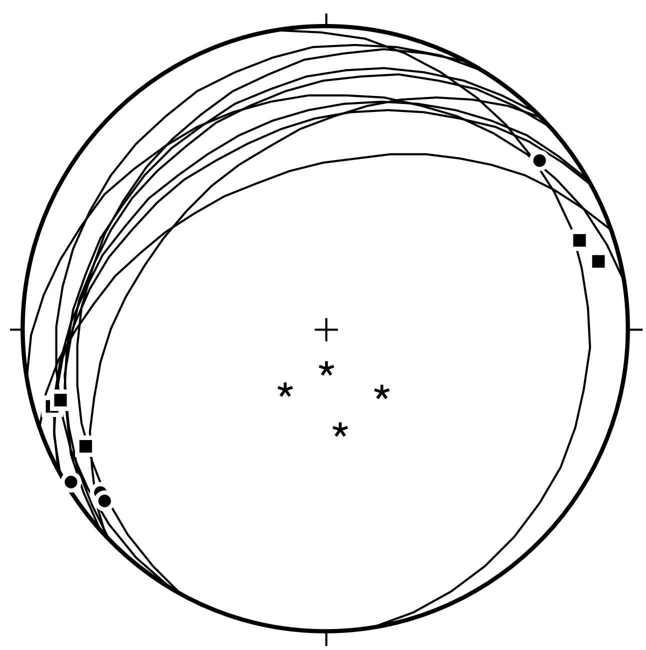

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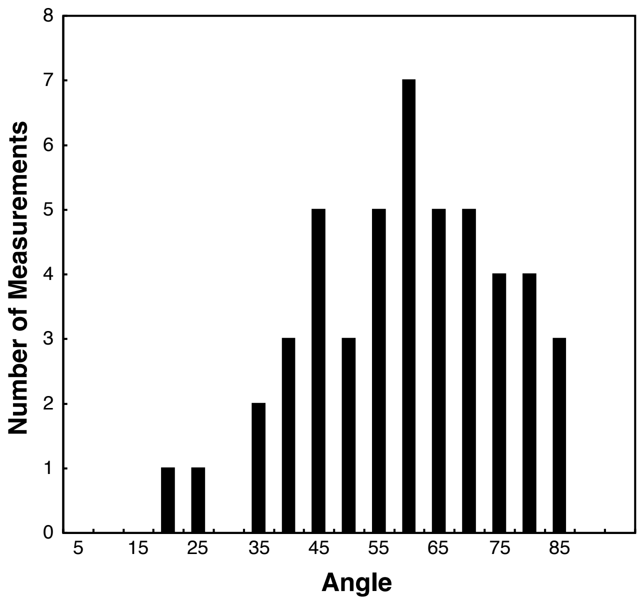

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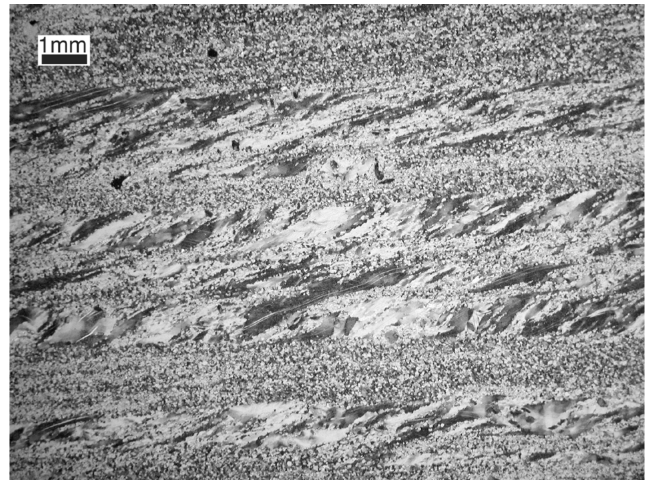

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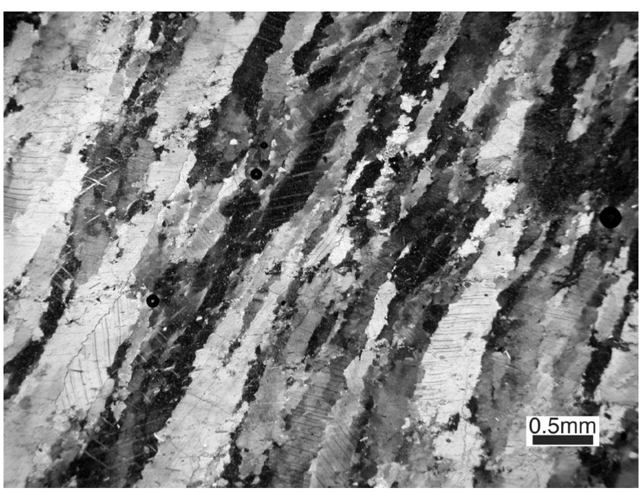

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