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Fluid percolation through quartz-rich lithologies: insight from the Coincident Site Lattice (CSL) Theory

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While connectivity of pore spaces created through suitable solid-solid-fluid dihedral angles is regarded as the major control on fluid movement in rocks, grain boundaries also constitute alternative pathways. This study aims to examine the existence of viable fluid pathways along grain boundaries in quartz-rich lithologies using the Coincident Site Lattice (CSL) theory, which is used extensively by metallurgists studying percolation behavior in metals.CSL theory describes the degree of coincidence across grain boundaries. Metallurgists enhance materials resistance to percolation by Grain Boundary Engineering (GBE) that involves increasing the proportion of CSL boundaries by deformation at room temperature, followed by annealing. This leads to an increase in the fraction of low $\sum$ CSL boundaries, $\sum$ being the operative parameter of CSL theory that defines the ratio between the area enclosed by a unit cell of the coincidence sites and the standard unit cell. It has been experimentally proven that an increase in the proportion of CSL boundaries increases the resistance of materials to corrosion by improving resistance of grain boundaries to percolation. Thus, higher the degree of coincidence, greater is the resistance to fluid percolation along the boundary. Studies on CSL boundaries have been defined only for the hexagonal, trigonal, orthorhombic and cubic systems, which may limit its applicability to multi-phase rocks. Since quartz is hexagonal and common in the Earth's upper crustal lithologies, assessment of fluid percolation through quartz aggregates has implications for fluid movement in the upper crust.

Fourteen quartzite samples, deformed at different metamorphic conditions varying from greenschist to upper amphibolite facies, have been used for this study. Electron Backscatter Diffraction (EBSD) analyses of the selected quartzites were carried out to determine CSL boundary distributions. The highest fraction of CSL boundaries ($\sum \leq 29$) has been observed in a quartzite which has been deformed at granulite-upper amphibolite conditions, and was subsequently annealed. The lowest CSL boundary fractions are present in quartzites deformed at greenschist and lower amphibolite conditions. There is good correlation between the fraction of CSL boundaries present and the grade of metamorphism, with the CSL fraction rising with increasing metamorphic grade. A striking increase in the fraction of CSL boundaries in quartzites deformed at upper amphibolite facies compared to those deformed at low to middle amphibolite facies conditions implies that CSL grain boundary formation is favored above a threshold temperature represented by the transition from the middle to upper amphibolite facies. Random high angle grain boundaries (RHAGBs) tend to counter the effect of CSL boundaries and provide pathways for fluids to percolate easily. Fractal analysis of RHAGBs connectivity was carried out to establish the degree of inter-connectivity of the RHAGB network in each quartzite. The RHAGBs connectivity decreases with increasing metamorphic grade and increasing fraction of CSL boundaries. Thus, quartzites deformed at higher temperature are more resistant to fluid percolation compared to ones deformed at lower temperature. This may provide an alternative explanation for the impermeability of quartz-rich granulite facies rocks to hydrous fluids.