den Brok, S.W.J., Morel, J., Zahid, M. (2001) Microstructural evolution of stressed solid-fluid interfaces: In-situ experimental observations using K-alum as a rock analogue material. 13th DRT conference Deformation Mechanisms, Rheology, and Tectonics, Noordwijkerhout, The Netherlands 2001. Abstract Volume Page 32.

Microstructural evolution of stressed solid-fluid interfaces: in-situ experimental observations using K-alum as a rock analogue material

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It has been shown in theory (e.g., Srolovitz 1986, Leroy and Heidug 1994) that the nominally flat surface of an elastically stressed solid is instable with respect to the growth of perturbations with wavelengths (λ) greater than a critical wavelength (λ_c). For $\lambda > \lambda_c$ the energy of the system may be lowered by the formation of a 'rough' surface of high stress valleys and low-stress ridges, thus providing a driving force for material transport, e.g. by diffusion through an aqueous solution from the valley to the ridge. In this case, the steady state geometry of the surface is theoretically controlled by a balance between the elastic strain energy driving an increase in the amplitude of the valley-and-ridge structure, and the surface energy driving a decrease in the amplitude, i.e. a smoothening of the valley-and-ridge structure.

According to Srolovitz (1986) the change in free energy (ΔF) in going from a flat surface to a surface consisting of valleys and ridges with an amplitude c and a wavelength λ may be approximated by: $\Delta F=(-c\lambda\sigma^2/4E)+2c\gamma$ where σ is the differential stress in the bulk, γ is the surface energy, and E is Youngs modulus. The critical wavelength (i.e. the wave length at $\Delta F=0$) is then equal to: $\lambda_c=8\gamma E/\sigma^2$.

For a typical rock-forming mineral such as quartz, with $\gamma \approx 0.5$ J/m^2 and $E \approx 50$ GPa, λ_c would be of the order of 20 µm for $\sigma=100$ MPa, and 5 µm for $\sigma=200$ MPa. Hence, provided that dissolution/precipitation processes take place fast enough, a microstructurally significant, stress-driven surface roughness could be developed by stressing the rocks within the elastic deformation field only. In principle, such a roughness would significantly affect solution/precipitation processes, such as, e.g., pressure solution, water-assisted cataclasis, and/or stress-driven recrystallisation. It is therefore important to investigate whether or not the theoretically predicted elastic strain induced roughness formation plays a role in natural rocks. Moreover, the wave length of the grain boundary roughness in natural rocks (e.g. of serrated grain boundaries) could perhaps be used to determine paleo-stress.

Using K-alum ($KAl[SO_4]_2 \cdot 12H_2O$) as a rock analogue material we experimentally investigated the effect of stress (elastic strain) on the microstructure of crystal surfaces in contact with an aqueous solution. Experiments were carried out at room temperature and at atmospheric pressure. Part of the experiments were carried out in-situ, i.e. under an optical microscope. Bulk differential surface parallel stresses fell in the range 3-15 MPa, i.e. well below the brittle yield strength of K-alum. We observed that regular arrays of macroscopically visible etch grooves developed on the originally smooth free surfaces when stressed. These grooves are typically 20-40 µm wide and 10-25 µm deep. The wavelength (20-60 µm) of the groove pattern decreases with increasing stress and depends on the crystallographic orientation of the crystal face. The grooves are oriented perpendicular to the compressive stress. They disappear soon after the stress is taken off. Part of the grooves changed position, i.e. moved over the stressed surface, during the experiments, at rates of several tens of µm´s per hour. At relatively high stresses the grooves developed into subritical fractures leading to cataclastic deformation of the surface.

The development of the grooves is well explained by the theory of Srolovitz (1986) and Leroy and Heidug (1994). The size and wavelength of the grooves is in reasonable agreement with values predicted by the theory. The formation of such instabilities could significantly affect the grain boundary structure in rocks, and have a major effect on dissolution and growth processes.

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