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Fibre growth in wet salt aggregates in a temperature gradient field

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ABSTRACT

Intense fibrosity develops in wet porous NaCl crystal aggregates (grain size 250–500 μm) held in a temperature (T) gradient field (0.5–4°C/mm) at temperatures between 20 and 50–60°C. *In situ* microscopic observation of the process shows that fibre growth is associated with T-gradient driven motion of tiny gas (air, water vapour) bubbles present in the saturated intercrystalline aqueous NaCl solution. Gas bubbles move through the intercrystalline pore fluid into the cold direction. They only move if they are next to an NaCl crystal; bubbles that are 'free' do not move. Each bubble is 'pushed' into the cold direction by a growing crystal fibre of the same diameter as the bubble itself. Fibres apparently grow due to oversaturation of the NaCl solution at the hot side of the gas bubble. Crystals dissolve at the cold side of the gas bubbles, apparently by undersaturation of the NaCl solution there. Thus, bubbles dissolve their way through NaCl-crystals and aggregates. Intense fibrosity develops within weeks.

KEYWORDS: fibre growth, salt aggregates, temperature gradient, NaCl, fracturing.

FIBROUS crystalline aggregates are common in deformed rocks where they serve as kinematic indicators of rock deformation. Development of such fibres is commonly assumed to be associated with local brittle deformation (fracturing), and opening of fluid-filled cracks (e.g. Ramsay, 1980; Ramsay and Huber, 1983; Urai *et al.*, 1991; Passchier and Trouw, 1996). Fibre growth then proceeds through growth at a fluid–liquid interface in the opening crack. Recently, however, intensely fibrous aggregates were found to develop in experiments without fracturing in wet NaCl and KCl aggregates, but within a temperature gradient field (Bons and Jessell, 1997). Individual crystal fibres obtained a length of several millimetres parallel to the temperature gradient in an aggregate of originally equidimensional grains of <100 μm in diameter. These experiments show that fibrous aggregates may form in non-fracturing rocks and represent a new and unexpected mechanism for the development of such aggregates in nature.

Bons and Jessell (1997) suggested three different possible mechanisms that might have been responsible for fibrous growth in their experiments: (i) local net precipitation of material transported in solution from the hot towards the cold end of the sample; (ii) anisotropic grain growth; and (iii) motion of fluid-filled grain boundaries in a way similar to the motion of fluid inclusions within temperature gradient fields. They remained uncertain, though, about what mechanism of these three exactly might have been responsible for the fibre growth that they observed. The difficulty was that the experiments were carried out in test tubes and microstructures studied after the experiment. The fibre growth could not be studied during the experiment. In order to gain more insight in the processes responsible for the fibrous growth in question, we decided to repeat their experiments *in situ*. We studied the growth of fibres in wet aggregates of NaCl grains (grain size 250–500 μm) in a temperature gradient (0.5–4°C/mm) between glass slides, *in situ*, under an optical microscope (Figs. 1, 2 and 3). The lower, hot end of the aggregates was held at 50–60°C and the upper, cold end at 18–22°C.

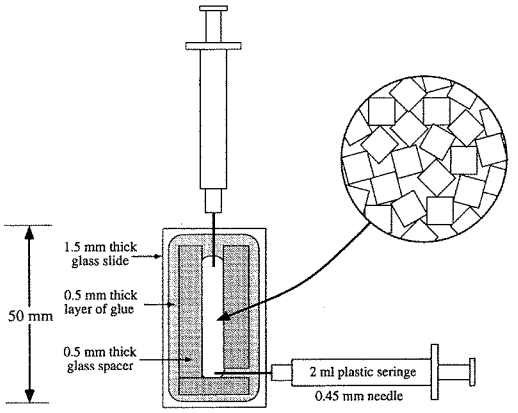

FIG. 1. Illustration of sample assemby used (to scale). NaCl crystals (pro analysi from Merck; 250–500 μm grain size) were inserted in the open space between two transparent glass slides that were glued together with Loctite 350 (UV-activated). The glass slides measure 1.8 × 48 × 28 mm; the open space measures typically 0.5 × 5 × 30 mm. Coverglass (0.5 mm thick) was used as a spacer. The crystal aggregate was carefully compacted manually to obtain a dense packing of the crystals. It was closed off from the outside by injection of glue in the opening. Two plastic 2 ml syringes with 0.45 mm diameter needles provided an in- and outlet to inject saturated NaCl solution. Needles were cut off after injection and closed by filling with glue.

Our experiments confirm the observation made by Bons and Jessell (1997) that intense fibrosity develops. However, we found in our experiments, that fibrosity develops due to motion of large numbers of gas (air, vapour) bubbles (10–500 μm in diameter) that were unintentionally present in the intercrystalline pore fluid. (Water was injected after evacuation of the air, but no special care had been taken to de-gassify the water. Large numbers of small gas bubbles grew by degassification of the added water at the beginning of the experiments.) On application of the temperature gradient, the gas bubbles started to move slowly through the aggregate, both through the pore fluid as well as through the crystals. They stopped moving as soon as application of the temperature gradient was stopped. Each gas bubble moving through the pore fluid left behind a crystal fibre,

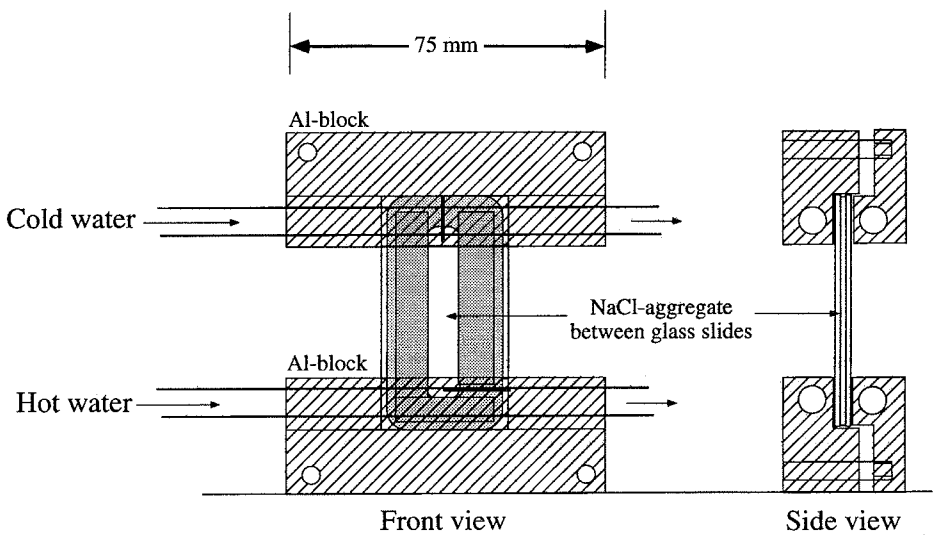

FIG. 2. Illustration of sample assembly (on scale) mounted between two aluminium blocks that have a different but constant temperature. Upper block was held at 18–22°C by circulation of cold tap water. Lower block was held at a constant temperature of 50 or 60°C (depending on experiment) by circulation of hot oil. Oil was kept at constant temperature using a Haake B3/DC5 temperature controller. The set-up is mounted on a microscope stage for microscopic observation.

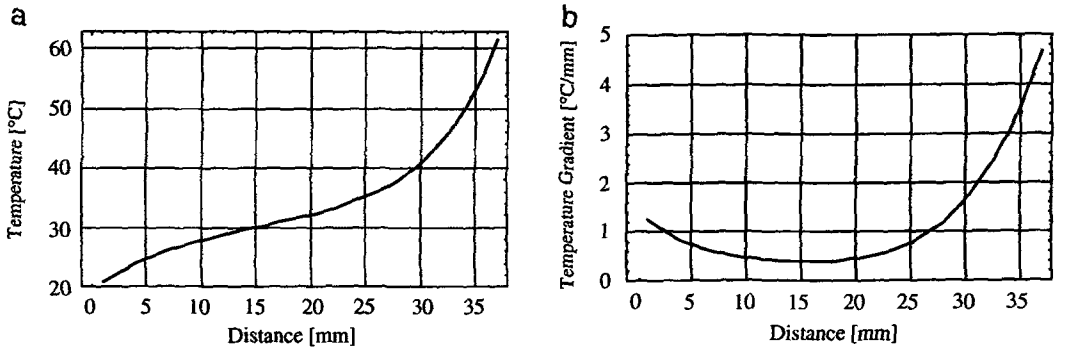

FIG. 3. (*a*) Typical temperature distribution, and (*b*) temperature gradient, measured from cold to hot parallel to the maximum gradient. Temperature distribution and gradient remained constant during an experiment.

that accurately tracked the bubble's motion (*Fig.* 4). Bubbles moved upwards, down the temperature gradient with velocities of the order of 2–10 mm/day. Some bubbles moved parallel to the temperature gradient, others moved at a small angle to the temperature gradient (less than

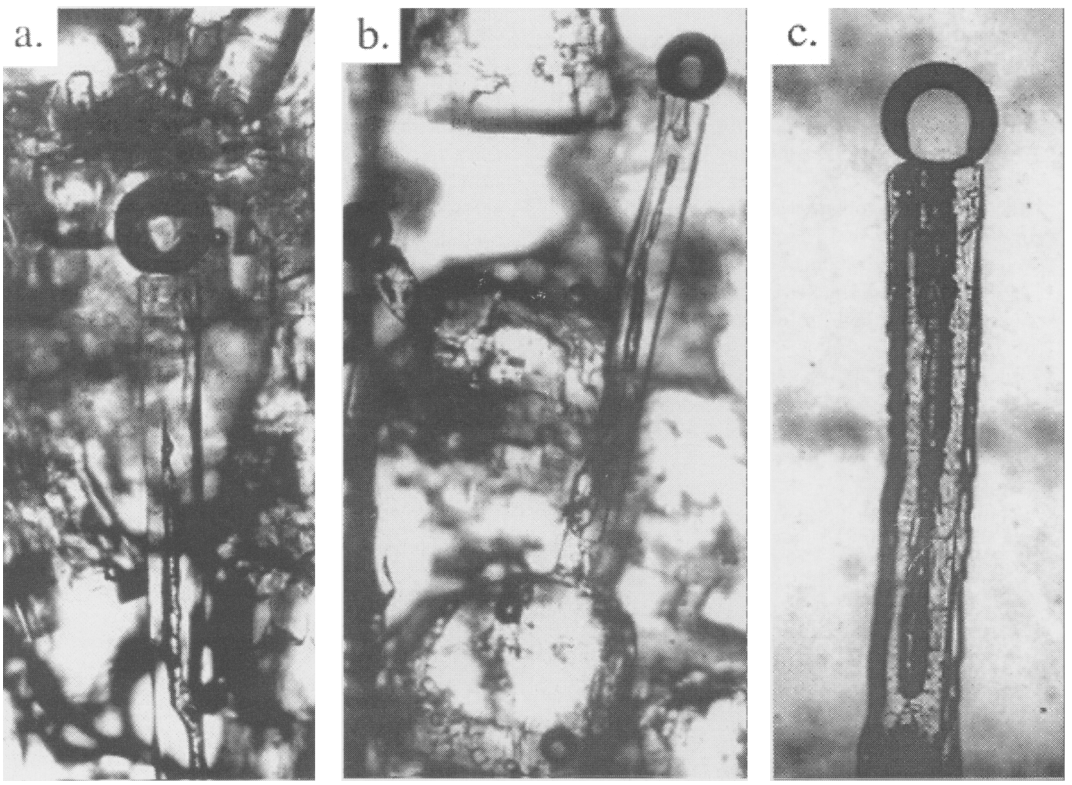

FIG. 4. *In situ* optical micrographs of NaCl crystal fibres grown in wet porous NaCl polycrystalline aggregate held in a temperature gradient of the order of 0.5–4°C/mm. Fibres tracked the motion of gas/vapour bubbles that moved into the cold direction. Cold above, hot below. Fibres grew parallel to (100) crystallographic direction. (*a*) Bubble diameter ~180 μm; (*b*) Bubble diameter ~140 μm; (*c*) Bubble diameter ~300 μm.

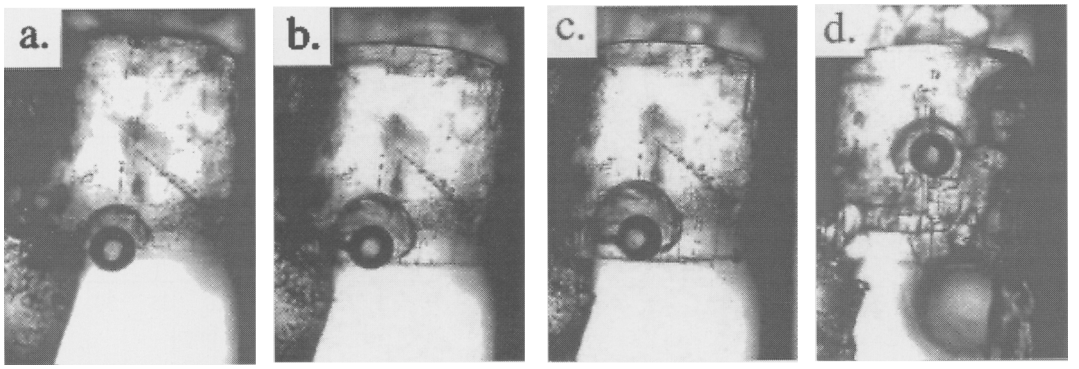

FIG. 5. Optical micrographs of gas/vapour bubble dissolving its way upwards into an NaCl crystal. Bubble moves towards the cold side of the aggregate. Note syntaxial overgrowth at lower, hot side of the NaCl crystal. Sides of the NaCl crystal are ~500 μm. (*a*) 4:50 hours after application of temperature gradient; (*b*) after 9:40 hours; (*c*) after 11:55 hours; (*d*) after 22:25 hours.

45°), but perpendicular to (100) or (110) of the NaCl crystal on which the fibre grew. Fibres grew syntaxially on their crystal substrate.

Gas bubbles right against the high-temperature side of NaCl crystals moved inwards into the crystals with some pore fluid (Figs. 5 and 6), and moved upwards, down the temperature gradient, at velocities of the order of 1 mm/day. In this way, some fibres tracked gas bubbles through the entire NaCl crystals. In other cases, bubbles became gas/liquid inclusions that were sealed inside a crystal. When gas/liquid inclusions moved out of the crystals they were tracked by fibres again (Fig. 7). The only gas bubbles to move were right against or inside NaCl crystals. Gas bubbles that were 'free' in the interstitial solution did not move. These apparently adhered onto the surface of the glass slides and would not move by buoancy alone.

The motion of gas/liquid inclusions within soluble salt crystals in a temperature gradient field, but without growth of fibres, was reported and studied by Wilcox (1969) and by Anthony and Cline (1972). No more recent reference was found. Anthony and Cline (1972) suggested that water is transported through the bubble in the vapour phase by evaporation/condensation. As a result, the salt solution will be supersaturated at the hot side and undersaturated at the cold side of the bubble, leading to precipitation and dissolution of salt. A gas–liquid surface tension gradient along the surface of the bubble (due to the gradient in concentration of the solid in solution) is assumed to drive a counterflow of water from the cold end to the hot end. Wilcox (1969) observed that small foreign particles moved rapidly around and spun on the surface of the bubbles, indicating significant motion of the fluid.

The mechanism proposed by Anthony and Cline (1972) for the motion of the gas/liquid inclusions through crystals can also explain fibrous growth. Due to continuous precipitation

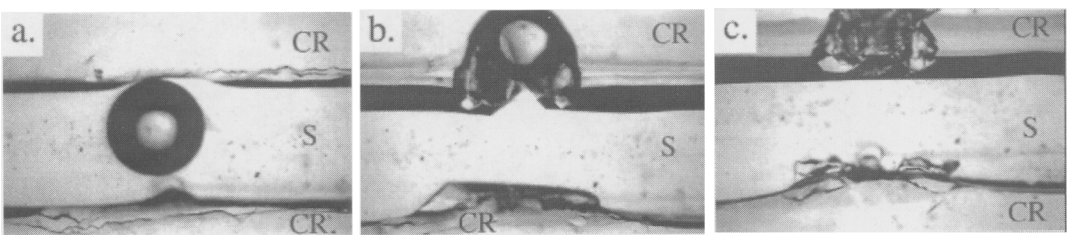

FIG. 6. Gas/vapour bubble dissolving its way into NaCl crystal at its upper, cold side. NaCl precipitates at the lower, hot side of the bubble. CR: crystalline material (NaCl), S: saturated NaCl solution. Bubble diameter ~700 μm. (*a*) 1:45 hours after application of temperature gradient; (*b*) after 23:20 hours; (*c*) after 72:00 hours.

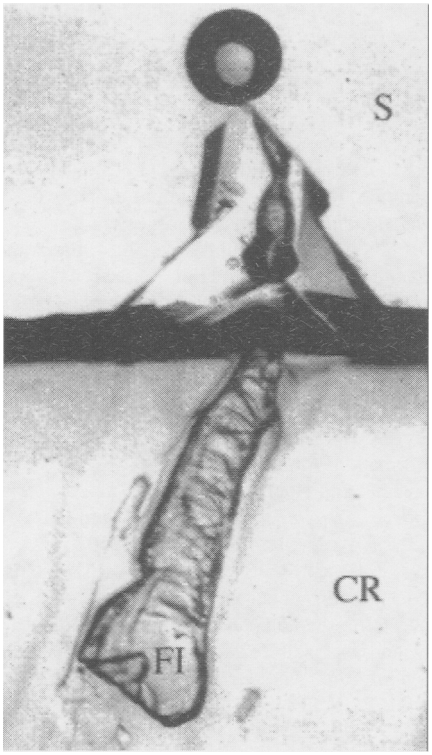

FIG. 7. Gas/vapour bubble that moved into the cold direction through the NaCl crystal (CR). It moved out of the crystal into the NaCl solution (S). The bubble was tracked by a tubular fluid inclusion (FI) and beginning crystal fibre. Cold is above, hot is below. Bubble diameter is ~180 μm.

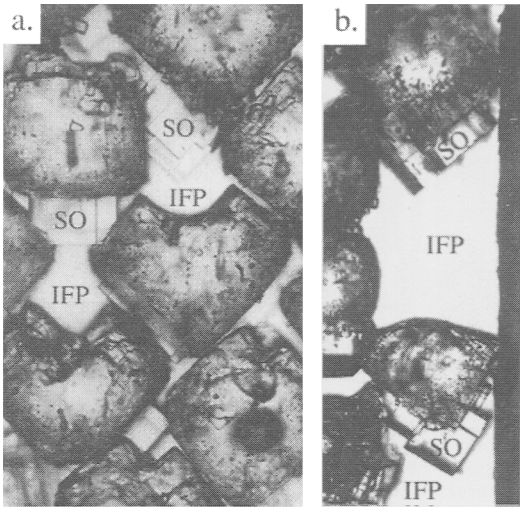

FIG. 8. Optical micrographs illustrating the motion of interstitial fluid pockets (IFP) towards the hot side of the aggregate. Crystals dissolve at their upper, cold side. Dissolved material is transported upwards into the cold direction towards the hot side of neighbour crystals where syntaxial overgrowth (SO) takes place. Size of the originally cubic NaCl crystals is ~500 μm.

of salt on the down-gradient side of a gas bubble a crystal fibre will develop there, and the bubble will migrate from the crystal substrate into the crystal growth direction by the fibre as more material precipitates. The fibre will tend to grow in a crystallographic preferred direction, i.e. parallel to (100) or (110), but the evaporation/condensation mechanism in the bubble will tend to guide the fibre down the temperature gradient.

In our experiments, fluid inclusions in the crystals and intercrystalline fluid pockets between the single crystals also moved, but towards the hot end of the sample, and at relatively low velocity (typically 0.1 mm/day). The motion of the intercrystalline fluid pockets contributed to the fibrous texture that developed in our samples, but to a lesser extent than migrating gas bubbles. The individual crystals surrounding the fluid pockets dissolved at their upper, colder side. The dissolved material was transported through the fluid in solution upwards to the neigbour grains, where it was precipitated at hot end of the grains (Fig. 8). The fluid pockets became progressively more elongate in shape, with apophyses pointing downwards to the hot side of the sample. The combined motion of the gas bubbles and the fluid pockets, albeit in an opposite direction, resulted in intensely fibrous aggregates developing within several days or weeks, depending on the temperature gradient (Fig. 9).

We found no evidence for an important contribution of the first two mechanisms suggested by Bons and Jessell (1997) to explain the observed fibrosity. The third mechanism, i.e. the motion of fluid-filled grain boundaries in a way similar to the motion of fluid inclusions within a temperature gradient field, clearly contributed to fibrosity in our experiments (Fig. 8). However, the most important contribution to fibrosity in our experiments was due to the motion of the gas bubbles through the aggregate. Part of the fibrosity in the experiments of Bons and Jessell (1997) may also be due to this mechanism. They might have been more succesful though, in suppressing gas bubble formation.

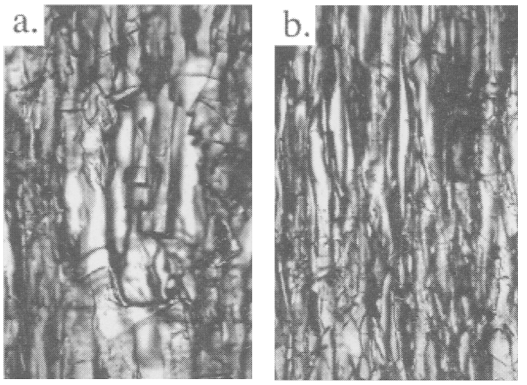

FIG. 9. Optical micrographs of wet porous NaCl aggregates illustrating intense fibrosity developed after several weeks in a temperature gradient of 0.5–4°C/mm. Cold above, hot below. Long side of pictures is ~2.5 mm.

It seems doubtful that fibre development by motion of gas bubbles in a temperature gradient field is a geologically important mechanism. It may be interesting to verify, though, whether it plays a role during and after storage of heat generating waste in natural rock salt deposits. Large temperature gradients are present across initially very wet and porous polycrystalline salt backfill that is used to seal the radioactive waste repositories (e.g. Spiers *et al.*, 1989). The intragranular fluid might contain abundant gas bubbles that would move away from the hot waste deposit, thus producing strong fibrosity and possibly high permeability in a direction away from the waste deposit.

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