Healing of fluid-filled microcracks

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ABSTRACT: The formation and subsequent healing of cracks and crack networks may control such diverse phenomena as the transport of radioactive materials in ceramics around nuclear reactors, the recurrence of earthquakes, and fluid migration in the Earth's crust. Although the process of crack opening has been widely studied for many years, the mechanism of crack healing remains controversial mainly because the driving forces controlling this process are not well understood. Here, we present the results of experiments in which a liquid was injected into an elastic gel to obtain penny-shaped cracks that were subsequently allowed to close and heal. Depending on the properties of the gel and the injected liquid, two modes of healing were obtained. In the first mode, the crack heals completely through a linear process. The unexpected second mode of healing is nonlinear and is characterized by a ”zipper-like” closure as the front moving along the crack margins may leave liquid inclusions trapped in the solid. This process is driven by the elastic rebound of the surrounding medium and the reduction in interfacial energy near the crack front. Our experiments reveal how linear arrays of fluid inclusions may be captured along preexisting fracture planes. This explains why previously fractured rocks may contain small amounts of fluids which permanently modify material properties.

1 INTRODUCTION

Crack healing is the closure of a fracture without any growth of external material in the fracture space (Laemmlein, 1929; Lemmleyn & Kliya, 1960; Bandyopadhyay and Roberts, 1976). Healing includes more than the process of fracture closure but, most importantly, also processes that reestablish the mechanical continuum that was broken during the fracture process. Healing is thus typically associated with the redistribution of material of the host mineral by diffusion processes and fluid/solid interaction. It has been proposed that the process is driven by reduction of the surface energy of the system and involves reestablishment of contact areas between the two surfaces of the open crack via the expulsion and/or redistribution of crack filling fluids (Nichols & Mullins, 1965a, 1965b; Nichols 1976). In this contribution we show that elastic and viscous forces can also play a key role.

In natural rocks, crack healing leaves arrays of fluid inclusions, with sizes of the order of 0.1 to 10 microns, permanently trapped at the former fracture plane. Crack healing in industrial materials has been studied for many years. For example, the rate of crack healing in ceramics used as protection shells around nuclear reactors is a major factor controlling the evolution of the pore space geometry, which in turn controls transport properties such as the permeability, of these materials (Wilson & Case, 1997; Wilson et al. 1997). A variety of crack healing experiments performed on ceramics (Roberts & Wrona, 1973), glasses (Wilson & Case, 1997) and crystals such as olivine (Wanamaker et al. 1990), calcite (Hickman & Evans, 1987) and quartz (Pecher, 1981; Smith & Evans, 1984; Brantley et al. 1990) show that the entrapment of fluid inclusions occurs via a two-step mechanism. In the first step, tubular fluid-filled voids form near the tip of the closing crack, and in the second step a necking process leads to the formation of strings of bubbles from these tubes. New cylinders may form as previously formed cylinders are undergoing necking. In calcite, for example, the final pattern consists of concentric strings of fluid inclusions with sizes that increase with increasing distance from the initial crack tip. In addition experiments show that crack healing is a thermally activated process and the rate is inversely dependent on crack aperture in calcite crystals (Hickman & Evans 1987). Disorderly surface roughness, and surface structure produced by processes such as conchoidal fracturing may lead to a complex pattern of solid-solid contacts during crack healing process. Such contacts may act as nucleation sites for subsequent crack healing process. In thin cracks, the growth of such contacts area may be the dominant healing mechanism.

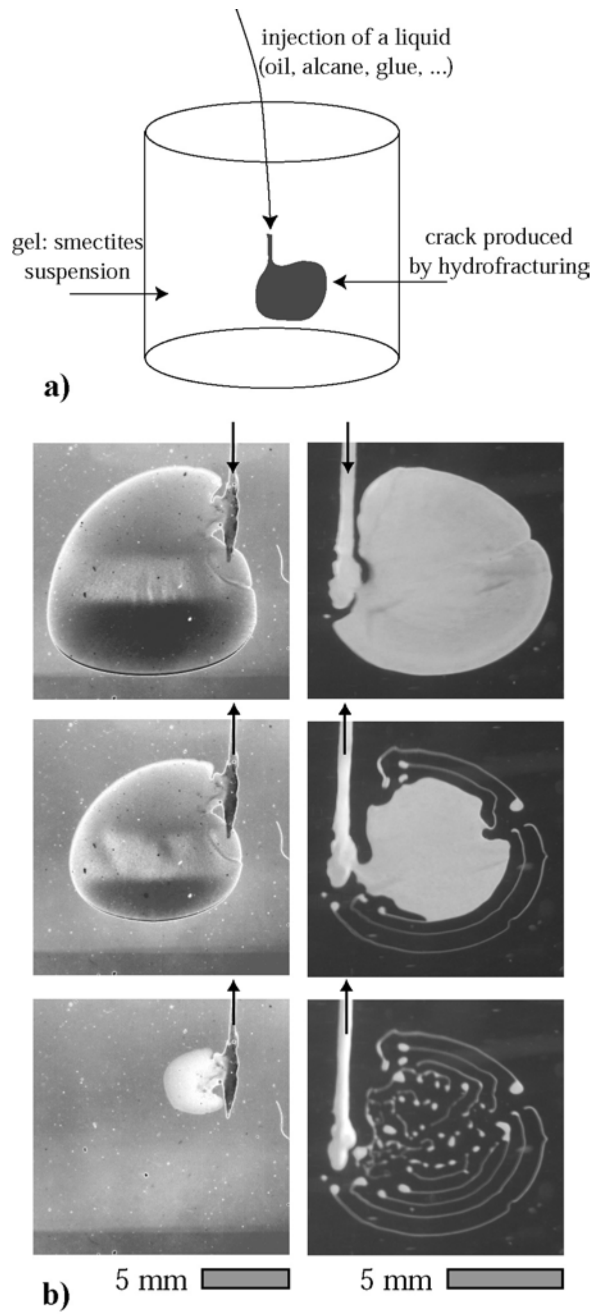

Figure 1. Hydrofracturing and two modes of healing. a) Injection of a liquid (epoxy glue, octane or heavy oil) into a Laponite® elastic gel. Penny-shaped cracks are produced by hydrofracture. Cracks are 1 to 3 centimeter wide and 100-300 micrometer thick. b) Experimentally healed crack in a brittle gel. In the top picture, a fluid was injected in a container filled with the gel. The pictures below show a time sequence. A 1 cm wide and 200 micron thick crack produced by hydrofracture formed after injection. A single crack plane emerged from the injection channel (arrow on the top picture). After several minutes, the fluid filling the crack had escaped through the injection hole (bottom picture). On the left side, the crack is completely healed whereas on the right side the healing front has left behind a network of concentric channels filled with fluid. The two experiments were performed with different fluids: paraffin oil on the left-hand-side, and a liquid epoxy with higher viscosity on the right-hand-side.

2 EXPERIMENTS WITH A SYNTHETIC GEL

Experiments reported in the literature do not follow the entire crack healing process, and a comprehensive understanding of crack healing have not been attained. For example, in isotropic solids, it is not known whether the distribution of bubbles and cylinders in an almost completely healed crack reflects the geometry of the initial surfaces or if it is controlled by a pattern forming process that acts during healing. Following the experimental studies on crystals (Wanamaker et al. 1990; Smith & Evans, 1984; Brantley et al. 1990) or on ceramics (Roberts & Wrona, 1973), there is still a controversy concerning which forces (surface tension forces or elastic forces) dominate crack healing and how the fluid inclusions pattern develops in time. Here we report the result of crack healing experiments in a brittle hectorite clay suspension (Laponite®), which produces a homogeneous gel when mixed with distilled water. We have chosen this material because it is transparent, it has a low elastic modulus such that the healing process is suitably fast at room temperature, and it has a viscous response under shear strain (Pignon et al. 1996). The samples are prepared by direct dispersion of the clay particles powder in distilled water. The mixture is stirred at high speed for 5 minutes. The pH of the sol ranged from 9 to 10. After a one-week storage at room temperature, a homogeneous transparent gel was formed. Depending on the concentration of clay particles (2 - 4.5 wt-%), the Young’s modulus of the gel varied between 500 and 2000 Pa (Mourchid et al. 1995, 1998). All the experiments presented here are with a 2000 Pa Young's modulus gel.

To initiate crack growth in the gel, a fluid was injected with a syringe (Fig. 1a) and induced hydraulic fracturing. When the tensile stress at the site of injection exceeds the tensile strength of the gel, a crack develops and propagates normal to the direction of the greatest tensile stress. During fracture propagation the strain energy is partitioned between a dissipative part and a reversible one. Plastic deformation at the crack tip dissipates energy whereas recoverable energy is stored in the system as new gel-liquid interfaces (surface energy) and elastic strain in the solid (DeGennes, 1987). The fluid injection formed pressurized penny-shaped cracks with diameters of the order of 1 cm and maximum aperture in the 100 and 300 micrometer range. To allow healing, the syringe is removed and the crack is left at rest. The hole left by the needle stays open and connects the fluid filling the crack to the outside. The fluid pressure is then released to the atmospheric pressure and the fluid contained in the crack is drained through the hole left by the needle as the fracture heals.

A variety of fluids with different surface tensions and viscosities were injected in the gel. These included a liquid light curing epoxy glue (Castall -- Fig. 1b, right side; Figs. 2a, 3a), hydrocarbon liquids (octane, paraffin oil -- Fig. 1b, left side), and motor oil (Diesel Oil BP 10W40 -- Fig. 2b). The epoxy is a low viscosity liquid that can be solidified by exposure to blue light for 30 seconds during any stage in the experiment. This provides a cast of the crack space and allows the geometry of the crack aperture to be accurately determined. In all the experiments, a high resolution charged coupled device (CCD) camera was used to record a series of images that could be analysed quantitatively and provided a record of the crack healing process.

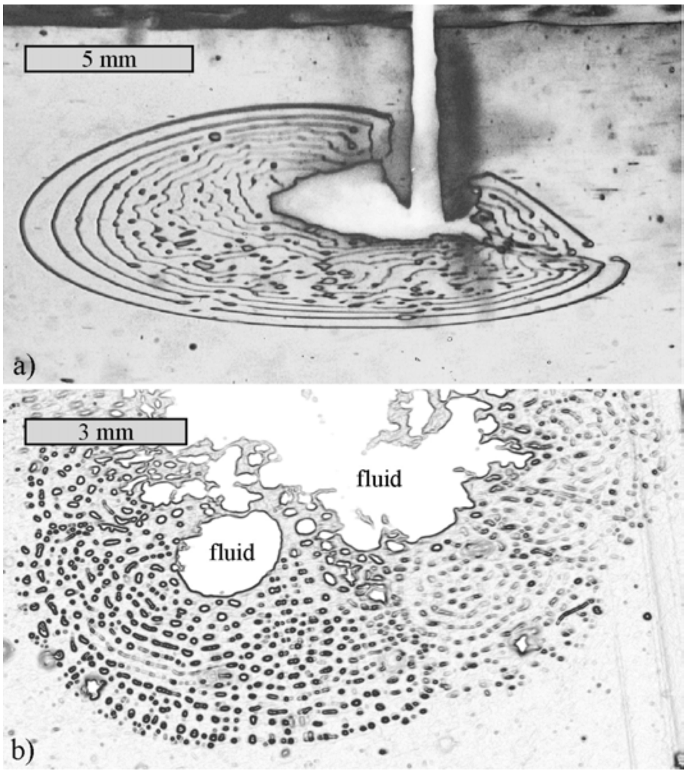

Figure 2. Geometry of healed cracks depending on the injected fluid. a) Healed crack originally filled with liquid epoxy glue showing the concentric channels of fluid trapped during the healing process. Some fluid is still trapped in the open portion of the crack. b) Healed crack originally filled with motor oil. With time the channels of trapped fluid become unstable and fragment into single spherical inclusions. Finally a concentric network of fluid inclusions pervades the healed region.

3 RESULTS

Depending on the properties of the materials used in the experiment, complete healing without any fluid trapping (Fig. 1b, left side) occurred; or the healing process produced a pattern of tubular fluid inclusions called "channels" (Fig. 1b, right side; Fig. 2a) that eventually necked off into series of spherical fluid inclusions (Fig. 2b). The formation of lines of bubbles via necking of the channels is the well-known Plateau-Rayleigh instability that is driven by reduction in the interfacial energy (Chandrasekhar, 1981; Faber, 1995). To our knowledge, the first step, the formation of annular channels near the crack tip, has not been explained in the literature.

By analysing epoxy casts of the crack, the early stage of healing can be studied. Using this approach, we have measured the crack tip geometry before channels had formed by using a white light interferometer. These experiments revealed that, during this early stage of healing, an annulus forms at the tip (Fig. 3b top). Combining this information with time-lapse videos establishes the following healing scenario. Initially, an annular channel develops at the crack periphery. There, an indentation forms at the periphery of the crack and grows until the annulus is penetrated and forms cylinders that are aligned along the crack circumference (Fig. 3b). This process appears to occur via the amplification of irregularities at the crack periphery. The annulus then detaches from the crack tip by a “zipper”-like mechanism (Figs. 3a, 3c) that removes fluid and closes the crack space in the region between the annulus and a new crack space of reduced size. A new annulus then forms along the periphery of the crack of reduced size. The process is repeated a number of times until almost the entire crack space has been converted into fluid inclusions. The thickness of the successive channels is constant, but the distance between the channels tends to decrease as healing progresses. Later, the channels become unstable to longitudinal thickness perturbations and fragment down to isolated spherical bubbles (Fig. 2b). Note that this late instability appears with motor oil (Fig. 2b) and not for epoxy (Fig. 2a).

4 DRIVING FORCES

During the experiments, the healed distance, that is the portion of the crack that has closed since the beginning of the experiment, can be determined as a function of time using the CCD camera images (Fig. 4). Depending on the nature of the injected liquid, two different crack healing processes and their kinetics were observed. On one hand, during complete continuous healing, the closing rate (the rate at which the healing distance increases) increased slowly. On the other hand, discontinuous healing by successive ovulations of a liquid cylinder leads to a healing rate that decreases slightly with time (Fig. 4). During closure, the evolution of the pressure field in the crack can be qualitatively described by the observation of the drainage of small particles initially present in the fluid. It appears to be closely related to the nature of the process. Our observations indicate that during continuous healing (Fig. 1b, left), the pressure field is radial near the crack tip so that the fluid moves directly from the crack tip to its center. In this case, a simple Darcy law can describe the drainage of the crack with a permeability that depends on the square of the crack aperture. During discontinuous healing, the fluid pressure gradient is tangential near the edge of the crack, and the fluid circulates parallel to the front of closure (Fig. 3b). Under these conditions surface tension plays a key role at the crack tip.

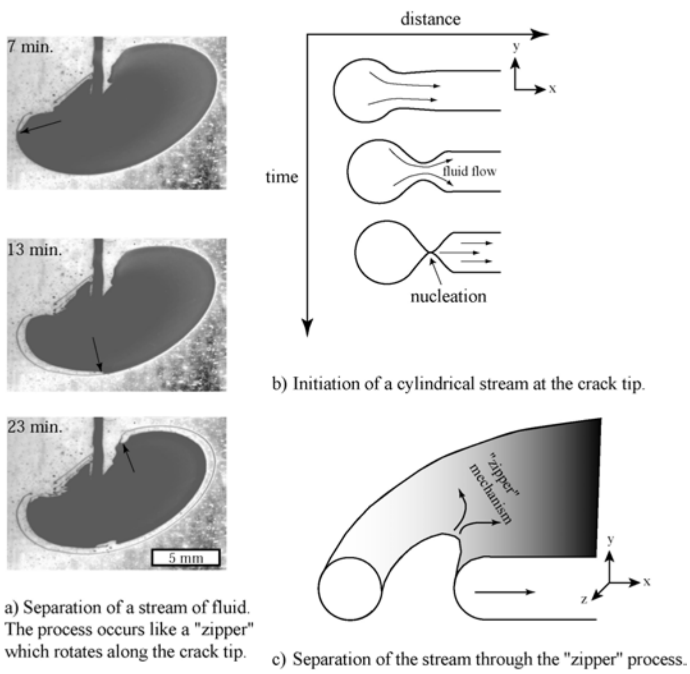

Figure 3. The “zipper” effect. a) Successive pictures of the detachment of a cylindrical fluid inclusion at the crack tip in an epoxy-filled crack. b) Sketch of the initiation of a cylinder at the crack tip. c) Then a channel of fluid separates. While the pressure gradient is radial in the open portion of the crack, it becomes tangential near the channel. The process is driven by surface tension and elastic energy released by the walls of the crack. The channels detach like a “ zipper” from the open part of the crack.

Under the conditions of the experiments, the forces involved in crack healing are surface tension forces at the solid-liquid interface, elastic and viscous forces in the gel and the viscous forces in the liquid. The coupled drive due to the elastic forces exerted by the gel on the crack-filling fluid and the non-uniform surface curvature at the crack tip, induces fluid transport and causes healing. Closure in these experiments occurs when the force that holds the fracture open is removed, i. e. when the internal fluid pressure is released. The pressure contribution induced by the elastic stress of the gel has a magnitude of the order of 10 Pa, whereas the pressure contribution due to surface tension is three orders of magnitude smaller. The initial pressure contribution due to elasticity, ΔP, is given by $\Delta P = wE/4c$ where w and c are respectively the crack thickness and width and E is the Young’s modulus of the elastic solid (Barenblatt, 1962). Using the modulus measured for such gels (Mourchid et al. 1995, 1998) and typical values for w and c from the experiments, an elastic contribution of 5 to 20 Pa is obtained, which is the main driving force that expels the fluid out of the crack. The relative importance of inertia forces associated with the fluid flow and the viscous forces can be estimated from the Reynolds number $R_e = U_0 L / \upsilon$ where U_0 is the velocity of healing, L is the thickness of the crack and υ is the kinematic viscosity. In our experiments, $U_0 \sim 10^{-5}$ m/s, $L \sim 10^{-5}$-10^{-4} m, and $\upsilon \sim 10^{-3}$-10^{-2} m^2/s. This leads to Reynolds numbers in the order of 10^{-7}, which indicates that inertial forces are negligible (Panton, 1992).

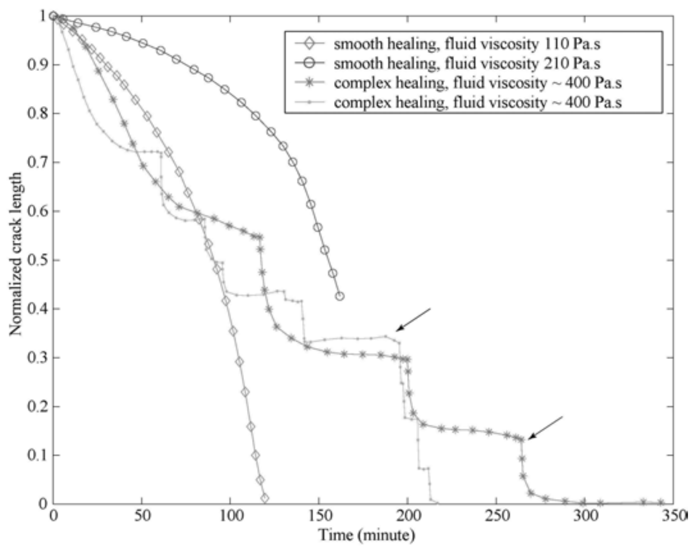

Figure 4. Crack healing rate as a function of the nature of the infilling liquid. The crack length is normalized to its initial maximum length. For paraffin oils (viscosity 120 and 210 Pa.s), the healing process is smooth (see also Fig. 1b, left side) and the rate of crack closure tends to increase with time. This is an example of an elastic dominated process. For light curing liquid epoxy (viscosity around 400 Pa.s), the healing process is discontinuous with velocity jumps (arrow) corresponding to the formation of cylindrical fluid inclusions such as those on Fig. 1b (right side). In this case, the process is controlled by surface tension. The final geometry of the healed crack is given on Fig. 2a.

The capillary number Ca of the experiment is $Ca = U_0 \eta / \sigma$, where U_0 is the velocity of the fluid ($U_0 \sim 10^{-5}$ m/s), η is the fluid viscosity ($\eta \sim 100$-400 Pa) and σ the surface tension ($\sigma \sim 10^{-3} \sim 10^{-2}$ N/m). This leads to capillary numbers in the order of 0.1 to 4. Low capillary healing is dominated by the coupling of elastic forces and surface tension. High capillary numbers are characterized by a balance between viscous flow and elastic forces on the crack walls.

Near the crack tip, elastic and viscous forces in the gel and tension at the gel-liquid interface compete. Unless the material has been plastically deformed, the surface tension and elastic forces should act together to close the crack. In addition, elastic forces should be dominant on long length scales and surface energy forces should be dominant on short length scales, near the crack tip. Reduction in curvature at the crack tip is controlled by surface tension. However the elasticity of the gel resists this process. On short length scales a characteristic distance at which the process switches from elastic-controlled to surface tension controlled is $d = \sigma/E$ where E is the Young modulus of the gel and σ the interfacial tension. If d is small enough such that elastic forces dominate, the healing process should remain continuous. When d is larger, the healing process near the crack tip becomes surface tension dominated and fluid inclusions can form. Our experimental conditions ($E \tilde{} 10^2\text{-}10^3\ N/m^2$, $\sigma \tilde{} 10^{-3}\text{-}10^{-2}\ N/m$) give a characteristic length $d \tilde{} 10^{-6}\text{-}10^{-4}\ m$. This length is in the same order as the crack thickness or to the mean spacing between the channels.

The tips of cracks have a high strain energy and therefore a larger chemical potential that the crack wall behind the crack tip. Therefore the crack tip is rapidly excavated by viscous forces under the influence of a high chemical potential of the gel relative to the fluid. There the chemical potential of the solid is lower relative to the fluid back along the crack wall, material is deposited to make a knob that becomes a "zipper". The fluid inclusion bubble stabilizes as a tunnel is created at the crack tip.

Mechanical crack closure and withdrawal of fluid from the closing crack is controlled by two sets of forces: (1) the tendency of the crack to close elastically, and (2) fluid pressure within the fracture with the tendency to resist closure. As the fluid is allowed to drain from the crack, fluid pressure is controlled by the viscous resistance of the fluid flowing out of the crack.

A different surface force effect results from the tendency to minimize the surface energy of the liquid/solid interface. This effect will tend to blunt the crack tip and lead to necking of the fluid layer in the crack. Blunting and necking necessarily involve either diffusive mass transfer or viscous/plastic flow of the solid. Elastic closure alone cannot result in necking of fluid inclusions or tip blunting because elastic fracture closure during slow draining of the fluid will maintain an equilibrium fracture shape. The equilibrium fracture shape is only controlled by the elastic properties of the solid (that are unlikely to change during the experiment) and the two sets of forces that control fracture opening as described above. Tip blunting and inclusion necking are a departure from the equilibrium crack shape, however, and require inelastic processes such as plastic/viscous flow. Based on the shape of fluid channels and inclusions, it is likely that inelastic processes also accompany necking.

5 CONCLUSIONS

This experiment shows that closure of a fluid-filled penny-shaped crack can be an unstable pattern forming process in the presence of surface curvature perturbations (Fig. 5). The final structure is an array of cylindrical and spherical inclusions. Estimation of the surface areas by image analysis indicates that the transformation from a planar crack to an array of spherical voids results in a reduction of the total interfacial energy of the system. Therefore, the healing process is driven by the interplay between the elastic energy stored in the gel when the crack opens, and released later, and the interfacial energy due to the formation of two solid/liquid surfaces. The formation of spherical inclusions implies that viscous deformation in the gel is important. Therefore the interplay between flow, plastic deformation, reduction of interfacial energy and reduction of elastic energy controls the pattern formation process. We do not have yet a full comprehensive model that explains the whole healing process, however this new highly nonlinear behavior is challenging for theoretical physicists dealing with coupled fluid mechanics and viscoelasticity.

The pattern forming instabilities studied in these experiments represent a good analogue model of crack healing in natural conditions as the final pattern looks like similar to what is observed in crystals (Hickman & Evans, 1987; Brantley et al. 1990). Such process indicates how fluid inclusions can be trapped in the rocks of the upper crust. The healing mechanism observed in elastic gels may also have important implications for earthquakes dynamics, the containment of nuclear waste and other processes in which fluids and cracks are known to play a key role. Fractures and microcracks play important roles in the transport of fluids in the Earth's crust. The mechanism of healing and the rates at which small cracks close are crucial issues in the development of a better understanding of the dynamics of fluid circulation in the crust (Moore et al. 1994). Small amount of free water trapped inside crystals also significantly modifies rock rheology and decreases the temperature of initial melting. The mechanism of fluid inclusions trapping observed in our experiments provides an explanation of how free fluids can remain in rocks under metamorphic conditions. Although the interfacial energy could be reduced by the coalescence of fluid inclusions or by escape of the trapped fluid, the formation of fluid inclusions is an essentially irreversible process under many conditions. Consequently, damaged porous solids that have been invaded by a liquid cannot usually recover their initial mechanical properties.

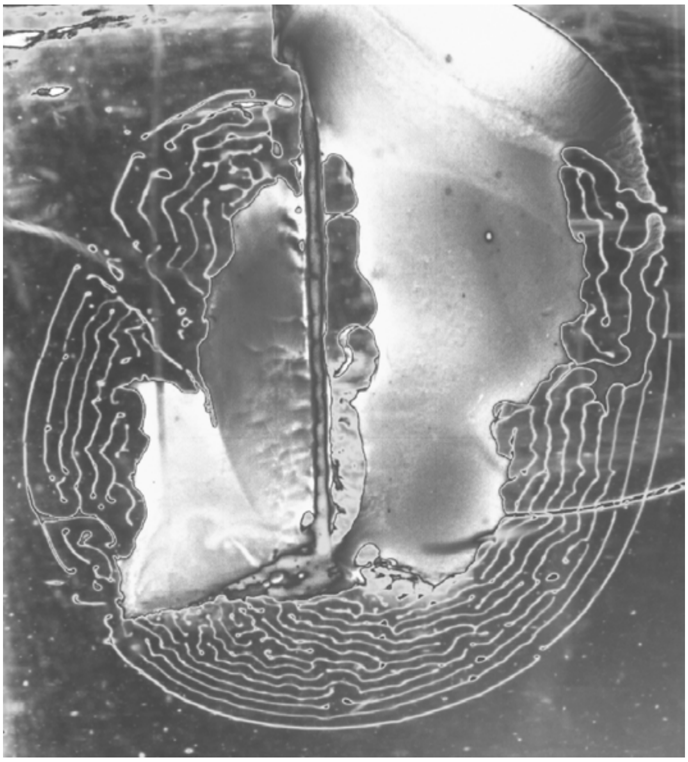

Figure 5. Fluid inclusion pattern of healed cracks.

6 ACKNOWLEDGEMENTS

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