ABSTRACT
Veins are common in Earth’s crust, and are formed by a wide range of processes, which lead to crystal growth in dilation sites. The first-order processes in vein formation have been identified, but it is much less clear how these can be diagnosed from field studies. In order to better understand the microstructural evolution during vein growth, we grew veins of analogue material [alum, KAl(SO₄)₂·12H₂O] in a transmitted-light cell from an advecting supersaturated fluid. Real-time observation shows the effects of flow rate and supersaturation on the evolving microstructure: (1) along-vein trends in growth rate caused by decreasing supersaturation, and (2) growth competition between clear crystals in the absence of nucleation and primary fluid inclusions. Although the overall trends in growth rate are in agreement with previous work, the local effects at the scale of individual grains reported here are less well understood; these new data form a basis for better interpretation of natural microstructures. To explore the possible effects of experimentally observed processes during vein growth, we simulate the growth kinetics of a quartz vein at various conditions of advective flow in Earth’s crust. Results show that in general the along-vein changes in growth rate occur at length scales much larger than a typical outcrop.

Keywords: vein, see-through experiments, syntaxial, sealing.

INTRODUCTION
The growth of crystals in dilation sites is of great importance in many fields of applied and basic earth science, because of the associated large changes in transport properties and rock strength (Blanpied et al., 1992; McCaig et al., 1995; Cox et al., 2001). A wide range of microscale processes has been identified, which, usually coupled, may contribute to the evolution of veins (Fisher and Branly, 1992; Renard et al., 2000). The resulting microstructures are correspondingly variable, ranging from blocky, elongate-blocky, to needle-like fibrous (Oliver and Bons, 2001). Isotopic signatures indicate that nutrients for vein growth can be supplied by long-range advective transport, or by diffusion in a local system (Kirschner et al., 1995; Elburg et al., 2002). Vein microstructure is interpreted to depend on parameters such as the degree of supersaturation, growth kinetics with different degrees of anisotropy, rates of local deformation, rates of transport, and relative rates of mineral growth vs. rates of vein wall separation. Therefore it is not surprising that diagnosis of the process of vein formation based on microscale structural and chemical information is inherently difficult. For example, the crack-seal mechanism has been diagnosed by distinctive microstructures (Ramsay, 1980), but this approach has subsequently been questioned (e.g., Wiltschko and Morse, 2001).

Most experimental studies on fracture sealing are based on dissolution, because the dissolution rate obeys the same first-order equation as the precipitation rate (Lasaga, 1998, p. 87; Ngwenya et al., 2000). This technique bypasses many experimental problems, but does not produce veins. Lee and coworkers (Lee et al., 1996; Lee and Morse, 1999) did pioneering experiments in which calcite was precipitated in a simulated fracture during advective flow. They identified and modeled the first-order growth process and predicted the kinetics of calcite vein formation in nature. However, the growth rates were too low to produce grains large enough to study the details of microstructural evolution. For such processes, analogue experiments are a useful alternative because they allow real-time observation at reasonable time scales (Streit and Cox, 2000).

In this study we present results from a transparent reaction cell, designed to simulate vein growth during advective transport. The model represents a simple isothermal system with flow of supersaturated fluid through a long, parallel-walled fracture. Fractures are sealed by epitaxial crystal growth on the fracture wall from a supersaturated solution. The results are applied to natural conditions and discussed in terms of information on the boundary conditions contained by the vein microstructure.

EXPERIMENTAL SETUP
The apparatus is placed under a microscope, and a solution of alum [KAl(SO₄)₂·12H₂O] is pumped at constant flow rate from the reservoir into the transparent reaction cell (Fig. 1). We used alum as an analogue material because of its well-known growth kinetics and high solubility (Mullin, 1993, p. 222–226). Flow rates were varied from 0.05 to 0.5 mL/h, which corresponds to an initial flow velocity of $1.3 \times 10^{-2}$ to $10.6 \times 10^{-5}$ m/s. Experimental results are applied to natural conditions and discussed in terms of information on the boundary conditions contained by the vein microstructure.
ments continued until the fracture was sealed and fluid flow stopped. The initial pressure gradient in the cell was 1.15 Pa/m. Pressure at the upstream end rose to locally much higher values of ~0.3 MPa shortly before the fracture was sealed and the experiment was stopped.

The reaction cell contains spacers and the model fracture. The simulated fracture is ~15 mm long, 2–3 mm wide, and 0.35 mm thick. It consists of fine-grained alum crystals that mimic the wall rock (Fig. 1).

Supersaturation of the fluid was induced by starting with a saturated solution in the reservoir; the temperature dropped by a few degrees Celsius between the reservoir and the reaction cell. This temperature reduction resulted in a supersaturation $S = c/c_{eq}$, where $c$ is the concentration in the supersaturated solution and $c_{eq}$ is the concentration in an equilibrium solution) of ~1.1, which is too low to induce nucleation from the solution. Temperature was controlled to $<\pm 0.05 ^\circ C$.

## RESULTS

In all experiments, epitaxial growth took place on the seed crystals along the entire wall length (Fig. 2). The grains are clear and without primary fluid inclusions.

Growth competition from the wall toward the vein center caused marked coarsening of the microstructure away from the vein margin. The final microstructure is a result of this growth competition, where most of the initially growing seed crystals were overgrown by a few grains (Fig. 3). The overall vein microstructure is elongate-blocky, similar to microstructures in syntaxial veins. Details of the growth-competition process are complex. Some grains grow much faster than their neighbors, overgrowing them and finally cutting them off from further solvent supply. Grains protruding into the channel at the initial stage do not necessarily survive during grain growth.

Crystallization and a resulting slight swelling of the matrix occur in the wall rock, as indicated by detailed observation of the high-resolution images. This swelling is interpreted to be due to pressure of crystallization, with a small part of the supersaturated fluid flowing in the matrix and causing growth of matrix crystals.

In all cases the average growth rate is highest at the inlet. To a first order, crystal-growth rates vary along the fracture as a function of flow rate and supersaturation. At a low flow rate of 0.085 mL/h, the growth rate decreases significantly toward the downstream end (Fig. 2A). The growth is more homogeneous along the vein if the flow rate is increased by a factor of 3, to 0.26 mL/h, keeping the supersaturation constant at the inlet (Fig. 2B).

The measured growth rates can be compared with results of a numerical simulation (Fig. 4), by using a finite-difference scheme, implementing the flow-velocity-dependent growth rate of alum (Mullin, 1993, p. 222–226; Hilgers and Urai, 2002). As found in earlier studies (Lee et al., 1996; Lee and Morse, 1999), there is a reasonable agreement between the trends in the experimental data and...
individual crystals (Fig. 4). This scatter is in-

strong scatter in growth rate at the scale of

ported in the literature.

ing lower flow velocity in the fracture and

flow passes through the matrix, with a result-

rection of transport. Information on the trans-

fracture and allows conclusions about the di-

are combined with the complex fluid flow

around the individual crystals.

Considering the final microstructures of our

experiments, we can ask the questions, Which

of these microstructures is diagnostic for the

single-pass fracture sealing of our experi-

ments, or, How can we use our results to im-

prove interpretation of natural vein microstructures?

First, there is a pronounced growth com-

petition, which results in a reduction of the

umber of actively growing grains with time. The

umber of grains versus time in our ex-

periments is shown in Figure 3. The details of

these trends have not been studied in any de-

tail, but may become useful microstructural tools to constrain conditions of vein growth after calibration with natural veins. All crystals grow into elongate-blocky morphologies with grain boundaries that are usually planar and irrational with respect to the surrounding crys-
tals. Occasionally there is a sharp change in
direction of a propagating grain boundary, asso-
ciated with the disappearance of a faster-
growing face. Upon sealing of the vein, crys-
tal faces disappear once opposing grains touch
(but their presence in the past might be de-
bected by cathodoluminescence microscopy).

An along-vein gradient in the amount of

precipitated material in a given time implies
transport (advective or diffusive) along the

fracture and allows conclusions about the di-

rection of transport. Information on the trans-

port process can only be derived if the sealed
zone is longer than can be expected from
short-range along-vein diffusional processes,
typically dominant to distances of ~10 cm.

APPLICATION TO QUARTZ VEINS

One of the characteristic features of along-

vein transport is decrease of growth rate along

the transport path. On the basis of this fact,
one may ask if such features could be observ-

able in nature and thus could be used as a tool
to investigate paleo-flow conditions. To test
this possible application, we use our numerical
model to calculate the growth rate of a quartz
vein. Because hydrodynamic effects on the
growth rate of quartz are unknown, we as-
sume that its growth kinetics are a function of
supersaturation only, described by a linear rate
law (Rimstidt and Barnes, 1980). Let us con-
sider a fracture with an aperture of 1 cm and
a length of 250 m, in which silica precipitates
from supersaturated fluid flowing at constant
flow rate and temperature. Following Rimstidt
and Barnes (1980), the mass precipitated in
each segment of the finite-difference grid is
dependent on the reactive surface area and the
supersaturation, the resulting growth rate be-
ing a function of the rate constant and the
transport mechanism. At $2 \times 10^{-8}$ m/s, a low-
er limit of flow velocities typical of crustal
fracture systems (Carson and Scretan, 1998;
Oliver, 2001), the initial pressure gradient re-
quired for flow along this 250-m-long fracture
is calculated by using the cubic law (Taylor,
1999) to be $10^{-4}$ Pa. Fluid pressure will in-
duce widening of the fracture if it becomes
larger than the normal stress, e.g., due to a
decrease in aperture.

Ross (1994) measured the concentration of
silica in pore waters in some hydrocarbon
fields and found a supersaturation $S$ of ~1.5
at 160–175 °C and 60–80 MPa. Thus, we con-
sider an $S = 1.1$–10 as reasonable for super-
saturated hydrothermal solutions and calculate
the growth rates of quartz in this fracture at
180 MPa fluid pressure and temperatures be-
tween 250 and 350 °C.

Similar to our experimental results and the
simulations of others (Lee et al., 1996; Dijk
and Berkowitz, 1998; Lee and Morse, 1999;
Giles et al., 2000), the growth rate decreases
along the fractures. More material precipitates
on the upstream side, and growth rates de-
crease toward the downstream end. The min-
imum lengths of a vein required to diagnose
sealing from advecting supersaturated solution
in an outcrop by a variation in material pre-
cipitated in the same time period along the
vein are ~943 and 196 m for calculations at
250 °C and 350 °C, respectively.

DISCUSSION

The relatively low supersaturation in our
experiments is reflected by the absence of nu-
cleation and primary fluid inclusions in the
vein microstructure. Because the formation of
primary fluid inclusions depends on the crys-
tal’s growth rate (Mullin, 1993, p. 261), we
propose that similar microstructures in natural
veins may be used to constrain paleosupersa-
turations. The system yielding steady flow is
a simple, experimentally accessible end mem-
ber of many different plumbing systems. Oth-
er transport regimes involve a constant pressure gradient or a constant flow velocity.

The aim of the experiments was a two-dimensional model of the growth process with visible microstructural evolution during fracture sealing. Vein growth stops when two opposing crystals touch and the fracture is sealed. In nature the solution will flow around locally sealed spots in three dimensions, allowing growth after the first contact between crystals growing from both sides. Therefore, longer sealed vein sections can be produced by advective flow, whereas in our experiments, the length of sealing is very limited.

Under these conditions, the vein microstructure will always become elongate-blocky, because the final remaining voids will form at the terminations of grain boundaries propagating from the vein wall into the center (e.g., in Figs. 2A and 2C). Syntaxial veins formed by this process will be neither fibrous nor produce grain boundaries that track the opening trajectory.

Advective mass transport is a major process in the crust. It is responsible for long-distance mass transfer and precipitation in many ore deposits (Heinrich et al., 1996; Cox et al., 2001). The large flow rates required for vein formation owing to the low solubility of most vein components are generally accepted (Wood and Walther, 1986; Heinrich et al., 1996). Consequently, naturally grown microstructures should contain information on the direction of transport. However, in many veins the diagnosis of advective flow from the microstructures grown is not straightforward. One problem is the change in rock composition and rock-grain size influencing the grown microstructure. Other factors are processes by which veins are completely sealed, i.e., crack collapse (Etheridge et al., 1984; Fisher and Brantley, 1992) or a slow compaction after the decay of fluid pressure.

Our calculations of quartz vein lengths predict that in outcrops sufficiently long, an along-vein trend of precipitated material in a given time could be used to diagnose advective flow in a single fracture. However, the length scale at which this decrease of along-vein growth rate is visible is too long to be observed in typical outcrops (Fig. 5). Thus, completely sealed microfracture arrays with small apertures frequently found in sandstones could have been formed by the advective-flow process considered in this paper.

The experiments presented have explored some of the basic characteristics of vein growth from an advecting fluid and contribute to the deduction of growth conditions from the microstructures observed. All veins were sealed at the inlet at variable conditions, the downstream growth rate decreasing more rapidly at low flow rates. More work is required for a full understanding. Expanding these experiments to include higher supersaturations and several stages of opening and sealing will allow investigation of the effect of these complexities on vein microstructure.

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