Impact of Surface Roughness on Evaporation in 2D Micromodels

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Abstract  Evaporation—a key process for water exchange between soil and atmosphere—is controlled by internal water fluxes and surface vapor fluxes. Recent studies demonstrated that the dynamics of the water flow in pore corners and thick-film flow on the rough pore-solid interface determine the fractality of the evaporation front. Based on our data, we estimate the film thickness and discover some interesting film-thinning effect with increasing temperature.

1. Introduction

Evaporation is an essential process in water exchange between the soil and the atmosphere. The evaporation rate from an initially saturated porous medium often exhibits a two-stage behavior: stage 1 is a constant rate period and stage 2 is a falling rate period (Brutsaert & Chen, 1995). Two factors determine the evaporation rate: continuous capillary flow and vapor diffusion. Capillary flow is the primary water transport mechanism, continuously delivered through the pore corners or surface cavities (Figure 1) to the evaporation front (Blunt & Scher, 1995; Hashemi et al., 1999; Lehmann et al., 2008; Sahimi, 2011). Capillary pumping (from the large pores to smaller pores) and continuous capillary flow are dominant in the early stage of evaporation (stage 1); vapor diffusion only becomes dominant in stage 2 when the continuous hydraulic connection is cut off and the remaining liquid clusters are isolated (Shahraeeni et al., 2012; Shokri et al., 2009).

Different types of capillary flows can occur depending on the morphology of the pore space and the geometric and physicochemical properties of the pore-solid interface. The geometrical structure of sharp corners enables a capillary flow along the corners, denoted as corner flow (CF; Blunt & Scher, 1995; see red arrows in Figures 1a and 1b), which has a smaller curvature radius and a larger capillary pressure than the duct flow (DF). For porous media with a rough inner surface, another type of capillary flow occurs inside the surface cavities. This capillary flow is driven by the high capillary pressure in the tiny cavities and generally carpets the entire rough surface; it is denoted as thick-film flow (TFF; Geistlinger et al., 2016; see the light-blue section in Figure 1b with flow direction indicated by a yellow arrow). Our study is in the pore scale (microns) with the pore diameters of the applied porous media (2D micromodel) mainly in the 200–600 µm range. The surface roughness of the glass-ceramic micromodel is within several microns (1–10 µm). In addition, an ultra-thin film flow can appear on surfaces for a different physicochemical reason. The nanoscale ultra-thin film flow is based on solid-liquid intermolecular adsorption due to the chemical surface properties, where flow in this film is driven by the disjoining pressure (de Gennes, 1985; Hu et al., 2020).
A study focuses on DF, CF, and TFF, determined by the geometric properties of the pore space and pore-solid interface and denoted as geometric capillary flow.

Evaporation at a slow rate is similar to the drainage process that both direct the displacement of a wetting phase by a non-wetting phase in porous media. During this displacement, the DF menisci recede into the pore channel with a curvature corresponding to the local capillary pressure; only the pore with the least capillary resistance is invaded at each time step. Evaporation leads to invasion percolation patterns, typical of drainage at low capillary numbers (Laurindo & Prat, 1996; Prat, 1995). Grate et al. (2010) and Zhang et al. (2011) showed CF during drainage using 2D silicon-pillar micromodels (smooth surfaces) and fluorescence microscopy. During the non-wetting phase invasion process, the wetting fluid did not completely recede, and the residual wetting liquid continually coated the silicon pillars in the CF form. The total interfacial area between the wetting and non-wetting phases (represented by the total perimeters of the coated pillars) was linearly related to the non-wetting phase saturation (Zhang et al., 2011). In the cross-sectional, confocal images in Grate et al. (2010), the retained wetting fluid was pinned throughout the silicon pillars’ height, while the fluorescence intensity was most intense at the bottom where the pillars met the pore channels’ horizontal surface. This result indicates the CF geometry as shown in Figure 1a. Dullien et al. (1989) showed the residual wetting fluid filling the cavities of the rough surface of etched beads using scanning electron microscopy (SEM) and microscopy. At the end of their drainage experiment, the etched beads’ surfaces appeared like they have been treated with several paint coats compared to the surface of unused etched beads. The bead surface cavities were filled with wetting fluid, forming a hydraulically conductive continuum of microchannels (TFF).

Three regions are formed in the porous medium during evaporation (Figure 2a): the fully saturated region, completely dry region, and partially wet film region. These three regions are segmented by the percolation front (DF front) and evaporation front (CF/TFF front). Except for a few isolated liquid clusters, the ducts in the film region are mainly occupied by vapor with a small amount of liquid retained in the corners or surface cavities (Figures 2b and 2c). In the drying experiment of Laurindo and Prat (1998), the top side of the network was an open surface, and the dye was introduced in the liquid feeding reservoir at the bottom after the steady state was reached. While the dye gradually invaded the micromodel through liquid transport, the color was observed in the saturated liquid region and trapped the isolated liquid clusters in the film region; these isolated liquid clusters were hydraulically interconnected to the liquid region by CF and TFF.

The three capillary flows’ hierarchy is determined by the driving water pressure $P_w$ (shown in Figure 1a), given by the negative capillary pressure $P_c$ ($P_c = P_g - P_w$, the gas phase pressure $P_g \geq 0$). According to the Young-Laplace law, $P_c \propto 1/r$, as the curvature radii of the three different flows obey the following relationship: $r_{DF} < r_{CF} < r_{TFF}$, the water pressure gradient shows a certain tendency ($P_{w,DF} > P_{w,CF} > P_{w,TFF}$), and water is transported from the saturated region toward the dry region through CF and TFF. When the film

Figure 1. Schematic models showing a capillary flow during evaporation, where the water phase is shown in blue, and the corner flow is indicated by red arrows: (a) Smooth surface capillary with corner and duct flows; the water-gas interfaces at positions 1, 2, and 3 are indicated by the blue solid, black-dashed, and red-dashed lines, respectively, in the cross-sectional image. Capillary pressure $P_c$ increases and water pressure $P_w$ decreases along the corner flow direction due to the decreasing curvature radius. (b) Rough surface capillary with duct, corner, and thick-film flows; the thick-film flow is shown in light blue with its flow direction indicated by a yellow arrow. The cross-sectional image of the red-dashed rectangle shows the thick-film flow filling the surface cavities and carpeting the entire rough surface.
region is full of vapor molecules, evaporation occurs intensively at the evaporation front (CF or TFF front tips), where the vapor concentration gradient is large and vapor diffusion is enhanced.

Lenormand and Zarcone (1984) have demonstrated the importance of TFF and that the timescale determines the relevant flow type during displacement. A capillary number $Ca = \mu_u/\sigma_{gw}$ (Blunt & Scher, 1995) was measured to present the relative effect of viscous forces versus capillary forces ($\mu_u$—dynamic viscosity of water, $\sigma_{gw}$—water surface tension, and $u_r$—the receding velocity of percolation front). At a high flow rate (capillary number: $Ca \geq 5 \times 10^{-1}$), the wetting fluid only exists in the duct (DF). At low capillary numbers ($Ca \leq 6 \times 10^{-1}$), CF and TFF occur with the same order of magnitude; for low flow rates ($Ca \leq 6 \times 10^{-2}$), TFF is the most significant water transport mechanism, facilitating the movement of wetting fluid throughout the network by flowing over the surface roughness, as the roughness term leads to a decrease in free energy (Zulfiqar et al., 2020). In our study, as the equivalent capillary numbers were in the range of $8 \times 10^{-1}$–$7 \times 10^{-2}$ ($u_r$ was derived by the experimental evaporation flux $Q_{ev}$ over the cross-sectional area $A$), we expect the TFF to be an effective water transport mechanism in our evaporation experiments.

Most studies have focused on the CF influence, while only a few studies have mentioned the impact of TFF on evaporation. Laurindo and Prat (1998) compared the drying curves for smooth and etched rough bead packs, finding that the etched beads’ drying rate was enhanced more by TFF than pure vapor diffusion in the smooth beads. Eijkel et al. (2005) conducted a theoretical study on the drying process in nanochannels, expecting the drying rate to increase by the combination of TFF and vapor diffusion relative to vapor diffusion alone. As few studies have investigated the impact of TFF on the evaporation process, we conducted evaporation experiments with 2D micromodels of different materials (silicon vs. glass ceramic). The silicon micromodels have a smooth inner surface where no TFF occurs, while the glass-ceramic micromodels contain a rough inner surface where TFF exists. As TFF has a lower water phase pressure than CF ($r_{TFF} > r_{CF}$), we expect that TFF will lead to a longer extension than CF. The longer film extension can maintain the medium surface partially wet for a longer period with a constant stage 1 rate. Additionally, we expect that, in stage 2, the long-extended TFF will deliver water to a higher position and closer to open surface than CF;

![Figure 2.](image)

Completely dry, partially wet, and fully saturated regions are segmented by the percolation (blue-dashed line) and evaporation (red-dashed line) fronts: (a) Experimental micromodel image after image processing, the evaporation front depth $x_e$, percolation front depth $x_p$, and corner flow/thick-film flow (TFF) extension $w$ are given by the geometrically averaged fronts; (b) schematic image of a smooth surface capillary with corner flow (yellow arrows), where the dry pore space in the partially wet region is full of vapor molecules (red dots); and (c) schematic image of a rough surface capillary with both corner and thick-film flow (light-blue film with the direction indicated by black arrows).
evaporation from the glass-ceramic micromodels will be more enhanced by TFF than that of the silicon micromodels. In summary, our working hypotheses about the impact of TFF on evaporation are:

1. TFF is an effective water transport mechanism
2. TFF will extend the stage 1 period
3. TFF will increase the evaporation rate during stage 2.

Owing to the angular geometry and rough surface of the real soil and sand grains, CF and TFF simultaneously coexist in natural porous media. To our best knowledge, no literature shows visualizations of CF and TFF during evaporation. Therefore, the main objective of our study is to provide adequate visualizations of CF and TFF, presenting a water transport between different capillary flows during evaporation using 2D micromodels.

2. Materials and Methods

2.1. Micromodel Fabrication

The 2D micromodels were designed with an identical size of 80 × 80 mm and a uniform inner channel depth of 0.3 mm. The microstructure named as MM5 (micromodel 5) is a quadratic lattice pore network (Figure 3a) containing 80 × 80 grain particles. The grain particles’ geometric centers are equidistant and connected by lognormal distributed throats with a throat width \( \mu = 400 \pm 62 \mu m \) (generated by MATLAB). The MM5 binary mask yields a network porosity of 0.52. This microstructure is similar to that used by Lenormand et al. (1988) to derive their famous phase diagram on fluid pattern formation; their mean \( 350-\mu m \) throat width corresponds to the \( 400-\mu m \) throat width of our MM5 microstructure.

Two types of materials were used for micromodel fabrication to differentiate the inner surface roughness. The silicon micromodel MM5-Si was constructed using inductively coupled plasma-deep reactive ion etching (ICP-DRIE) technology (Chomsurin & Werth, 2003; Küchler et al., 2003; Willingham et al., 2008; Zuo et al., 2013), providing a high edge steepness and the absolute smoothness of the inner surface (Figure 3b). The Si microstructure was covered by a thin SiO\(_2\) layer to achieve a similar wettability compared to siliceous materials (for details, see Geistlinger et al., 2019).

This etching technique minimized the underetching to a 1.2° deviation from the vertical line and maximal 10-\( \mu m \) deviation from the horizontal bottom line, reducing the corner rounding effects, which is the most compared to other etching methods (Vorhauer et al., 2015). Three sides of the micromodel were sealed, while the fourth side was left as an open boundary. For more details on the fabrication of the silicon micromodel, see Geistlinger et al. (2019).

The glass-ceramic micromodel MM5-glass was constructed by a photolithographic etching method, providing an accuracy in the order of 10 \( \mu m \) considering the sharpness of the grain wall. The solid, photosensitive glass-ceramic FOTURAN material (Figure 3c; Schott GmbH) has a rough surface with a mean roughness of 1–3 \( \mu m \), as provided by the manufacturer (Invenios Europe GmbH). The mean surface height \( R_h = 1 \mu m \) and the wavelength parameter \( D = 2 \mu m \) were the estimated surface roughness parameters.
indicating the surface periodicity (Geistlinger et al., 2016). The chemically etched glass micromodels were covered with a plain glass plate by a thermal diffusion bonding (500°C). After the micromodel construction, the etched structures were flushed with isopropanol and water and dried at 200°C.

Again, we emphasize that both MM5-Si and MM5-glass have the identical quadratic lattice microstructure (MM5) shown in Figure 3a, but different inner surface roughness shown in Figures 3b and 3c (due to a lack of the SEM image of MM5-glass from the manufacturer, we indicate the rough pore surface by another micromodel, which was fabricated in the same glass-ceramic material as MM5-glass in Figure 3c).

2.2. Methodology and Experimental Setup

2.2.1. Evaporation Experiment With DSLR Camera

Evaporation experiments were conducted with both MM5-glass and MM5-Si under different thermal conditions (room temperature (RT; 21–23°C), 42°C, and 61°C). Before each experiment, the micromodel was saturated with deionized water in vacuum for 2 h with an evacuator. For the above-ambient-temperature experiments, the micromodel was mounted on a metal heating plate (see Figure 4), while a temperature controller was attached to the heating plate to ensure a constant and uniform thermal field over it. During evaporation, the micromodel was vertically positioned with the open surfaces on the top. The evaporation process was continuously monitored using a high-resolution digital single-lens reflex camera (Canon 5D Mark IV, lens: Canon EF 100 mm F2.8L Marco IS USM) with a resolution of 6720 × 4480 pixels. The
total mass of the micromodel and heating plate was continuously measured using a high-precision digital balance (Sartorius Secura 1103-1S, ±0.001 g) and automatically recorded every 10 s using DasyLab. The laboratory’s relative humidity (RH) and ambient temperature during the experiments were recorded using a data logger (dataTaker DT80 Series 2). More details can be found in Geistlinger et al. (2019).

2.2.2. Evaporation Experiment With Fluorescence Microscopy

To better visualize the capillary flows during evaporation, horizontal experiments were also conducted under a fluorescence microscope (Leica Leitz DMRB, objective lens of Nikon CFI Plan Apochromat Lambda 2X), using a fluorescent dye (uranine) for the water phase. An SLR camera (Canon EOS 7D, spatial resolution: 5184 × 3456 pixels) with a 100-mm Marco lens (Canon EF 100-mm F2.8 USM Marco lens) and an LM Digital SLR adapter (LMscope C-Mount) were used. After each experiment, micromodels were flushed using water 10 times to remove the residual uranine and dried at 60°C for one week to obtain a reproducible initial condition.

2.3. Image Processing

The pore size distribution (PSD) and fluid patterns were analyzed using Fiji/ImageJ (Schindelin et al., 2012). The PSD was derived using the Fiji plugin “Local Thickness” (Dougherty & Kunzelmann, 2007), which inserts maximum-inscribed spheres (representing the pore size) into the pores (Figure 5b). The best-fit curve for MM5 (lognormal distribution, red curve) yielded a 197-μm mean pore radius with a 55-μm standard deviation (Figure 5a).

The grain mask binary image (developed for micromodel etching) must be registered onto the raw fluid pattern image for preprocessing. The “Big Warp” plugin (Bogovic et al., 2016), where arbitrary landmarks can be selected on the mask image and fluid pattern image for deformable image alignment, was used. For the MM5-Si images, completely dry pores and water-saturated pores are characterized by the bright edges of grain particles (Figure 9), because the light irradiating at the grain edges reflects back in the same way. In the partially wet pores, the CF slope eliminates this light artifact and presents a dark edge. Thus, the grain perimeters’ high gray values (black 0, white 255) are essential parameters for segmentation. For MM5-glass, the completely dry pores present a bluish background, whereas the wet pores are brownish (Figure 10). White stripes are observed in the partial wet pores where CF and TFF exist. The blue channel image was used for thresholding. As the fluid pattern image exhibits a radial illumination drift caused by the circular lighting source, it is difficult to choose a unique threshold value that applies to the entire image. Instead, we conduct image processing for a specific section of the image, which vertically includes the dry, saturated and whole film regions and horizontally covers the whole width of the micromodel. The chosen section is further divided into five small windows, so that each window can run a threshold depending on the local illumination condition (see Supporting Information S1).
The residual water saturation $S_w$ was derived by calculating the ratio: the total pixels of water-saturated phase (from the processed image) over the total pore space pixels (from the mask image). The evaporation mass loss $\Delta m$ is given by:

$$\Delta m = M_0 \cdot (1 - S_w),$$

where $M_0$ is the micromodel’s total water capacity.

The evaporation front is given by the upper boundary of the wet region (including the water-saturated and film regions), while the percolation front is given by the upper boundary of the water-saturated region. Given the $Y$ coordinate of each pixel on the evaporation/percolation front, we obtain the mean evaporation ($x_i$)/percolation ($x_p$) front depth (Figure 2a). The CF/TFF extension $\omega$ (Figure 2a) is given by the distance between the two averaged fronts.

### 3. Theoretical Background and Physical Mechanisms

#### 3.1. Full Duct Flow Versus Corner Flow and Thick-Film Flow

The standard model of evaporation (isolated pore-evaporation model = IPE model; see Figure 1a in Geistlinger et al., 2019; and Figure 5 in Shahraeeni et al., 2012) assumes that evaporation starts at the large pore menisci, and “capillary pumping” from these large pores to smaller pores determines the early stage of evaporation (stage 1). The key parameter is the pore size distribution or more specifically the hydraulic connected region of the PSD. The PSD of the micromodels (Figure 5a) corresponds to that of coarse sand with a mean pore radius of approximately 100−200 μm (Geistlinger & Zulfiqar, 2020), which causes a high hydraulic conductivity. Hence, viscous forces can be neglected in case of low-demand evaporation rate $(Ca \approx 10^{-7}−10^{-8}$; see Figure 1b in Shahraeeni et al., 2012) and the stage 1-behavior is controlled by water redistribution along the (water pressure) gradient from large to small pores. Since both micromodels MM5-Si and MM5-glass possess an identical pore structure, they should exhibit a similar stage 1 behavior as shown below in Figure 11. If the menisci have receded into the pores, water flow still exists within the pore corners and along the rough pore surfaces. Both water flows, which will be discussed next, are dependent on geometric properties and wettability of the inner pore-solid interface.

#### 3.2. Corner Flow Dynamics

To describe the corner flow dynamics during the whole evaporation process (stage 1 and stage 2), Yiotis et al. (2012) proposed a capillary-bundle model of disconnected effective single-square capillaries (SSC-model). This model was successfully applied to the evaporation process of the MM5-Si-micromodel (Geistlinger et al., 2019). A cross-sectional schematic at the open boundary ($x = x_0$) of such an effective square capillary is shown in Figure 6b. The vertical cross-sectional schematic along the diagonal cross-section of the capillary (red-dashed rectangle in Figure 6a) is displayed in Figure 6c, indicating the water-air interface movement from $t_0$ to $t_1$. Before the evaporation starts ($t_0$), the capillary is initially saturated with a meniscus $\Sigma$, some water remains in the sharp corners, and the water-air interface at the open boundary is marked by the solid black curves in Figure 6b. During the evaporation, the duct flow (DF) meniscus recedes into the pore channel with a stable curvature, and the corner flow (CF) extends from the water meniscus to the open surface. Meanwhile, CF recedes into the corners with decreasing thickness and increasing curvatures (red arrow in Figure 6b) until only the CF front tip is left at the open surface (e.g., $t_2$ in Figure 6c). For CF, the curvature along the $x$ direction can be neglected compared to the curvature in the $y$ direction (perpendicular to the $x$ direction). Therefore, the capillary pressure at the CF root (transition zone from DF into CF; e.g., $x_1(t_1)$, $x_2(t_2)$, and $x_3(t_3)$ in Figure 6c) equals the capillary pressure at the DF meniscus given by the Young-Laplace equation:

$$P_c(x) = P_c(x_1(t_1)) = P_c(x_2(t_2)) = P_c(x_3(t_3)) = \sigma_{gw} \cdot H(r_x, r_y) \approx \frac{\sigma_{gw}}{r_x},$$

where $\sigma_{gw}$ [N/m] denotes the water-air interfacial tension and $H$ [1/m] the curvature. The terms $r_x$ and $r_y$ are the curvature radii at the CF root ($x = -x_p$) in the principal directions $x$ and $y$, respectively (Geistlinger et al., 2019).
The radius \( r_{\text{E}} \) (Dong & Chatzis, 1995; Legait, 1983) is defined as (with contact angle \( \theta \)):

\[
F(\theta) = \frac{\theta - \frac{\pi}{4} + \cos^2 \theta - \sin \theta \cos \theta}{\cos \theta - \sqrt{\frac{\pi}{4} - \theta + \sin \theta \cos \theta}}, \quad \theta < \pi/4
\]

where \( r_{\text{E}} \) is half of the mean capillary width (Figure 6b). For a contact angle \( \theta = 0^\circ \), \( F(0) = 1 + \sqrt{\pi/2} \approx 1.886 \), the capillary pressure at the CF root is \( P_c = 1.886 \sigma_{gw}/r_{\text{E}} \), and for a typical contact angle of the oxidized Si surface with \( \theta \approx 40^\circ \) (Geistlinger et al., 2019), the capillary pressure is reduced by approximately 30%. We emphasize that \( F(\theta) \) becomes undefined (= 0/0) at the critical contact angle of 45° and consequently, CF is not possible.

Equilibrium between capillary pressure \( P_c \), air pressure \( P_a \), and water pressure \( P_w \) is established at the water-air interface in steady state: \( P_c = P_a - P_w \), which yields the water pressure \( P_w = -P_c \). At the CF front tip, the water pressure is approximated by \( P_w = -\sigma_{gw}/r_c \) with curvature radius \( r_c \). The water pressure gradient \( \Delta P_w = \sigma_{gw}/r_c = 1.886 \sigma_{gw}/h_0 \) (Figure 6c) drives the continuous transport of water from the DF toward the open surface through the four CF paths.

Owing to the exchange of water-air molecules along the entire water-air interface, the vapor concentration is nearly saturated within the CF region. When further water supply from DF was hindered (\( t_3 \) in Figure 6c), the CF front tip becomes dry, and the CF can no longer extend to the open surface, which is defined as the hydraulic detachment. Consequently, a completely dry region is formed above the CF front. The vapor concentration undergoes a decline from the saturated vapor concentration \( C_{sv} \) in the CF region to the ambient vapor concentration \( C_{av} \) at the boundary layer \( x = \delta \). The averaged evaporation rate can be described by the diffusion flux based on Fick’s first law.
\[ \langle Q_{ev} \rangle = A_y \left( J_{i, diff} \right) = \frac{\Delta C}{\Delta x} = \frac{-A_y D_e^{eff} C_x - C_y}{\delta + |x|}, \tag{3} \]

where \( A_y \) denotes the cross-sectional area of the gas phase, \( J_{i, diff} \) is the diffusion flux, \( D_e^{eff} \) is the effective gas-phase diffusion coefficient of the water molecules, and \(|x|\) is the distance between the evaporation front and open surface. In stage 1, \(|x| = 0\), the evaporation rate remains constant. After the CF detaches away from the open surface, the evaporation rate drops owing to the decreasing vapor concentration gradient, characterized as stage 2.

Physically, one expects that viscous forces/dissipation control the CF during stage 2, because of the small effective cross section in the upper section near the evaporation front (Figure 6). Besides the geometry of the pore channels, also the physicochemical state of the pore-solid surface (wettability and contact angle) has a significant impact on the CF and implicitly on the evaporation rate. Since the measured contact angle on oxidized Si surfaces is approximately 40°, it will reduce CF significantly compared to complete wetting (Geistlinger et al., 2019).

### 3.3. Thick-Film Flow Dynamics

In addition to corner flow, the micromodel MM5-glass also has the thick-film flow (TFF) with water retained in the surface cavities during evaporation. As a film carpeting the rough surface, TFF has a water-air interface at the film front with a curvature radius \( r_f \) along \( x \) direction (\( r_f \ll r_0 \)); curvature along \( y \) direction can be neglected. Water is transported from DF toward TFF under the water pressure gradient between the DF meniscus \( P_{w,DF} \) and TFF front \( P_{w,f} \), according to the following equation:

\[ \Delta P_w = P_{w,DF} - P_{w,f} = \frac{\sigma_{gw}}{r_f} - \frac{1.886 \sigma_{gw}}{r_0}. \tag{4} \]

From \( t_1 \) to \( t_2 \) (CF detachment time), the TFF carpets the entire rough surface of the partially wet pore (Figure 7c). Instead of the four corners in MM5-Si, the interconnected CF and TFF cover the entire perimeter of the open capillary surface as evaporation exits the MM5-glass (Figure 7b). CF and TFF directly contribute to the evaporation flux: \( Q_{ev} = Q_c + Q_f \). The capillary inner surface area covered by DF and CF is gradually replaced by TFF during the CF shrinking and DF receding processes. Owing to the water pressure gradient: \( P_{w,DF} > P_{w,CF} > P_{w,TFF} \), after water supply from DF vanishes \( (t_1) \), TFF can still be supplied by CF and maintains the hydraulic connection to the open surface, prolonging stage 1. Therefore, TFF detachment from the open surface, defines the end of stage 1 for MM5-glass.

### 4. Results and Discussion

#### 4.1. Corner Flow and Thick-Film Flow Visualization in a Pore Channel

The time-sequence images in Figures 8a–8d show the CF development in an MM5-Si pore channel during experiment 3 (61°C; Table 1). The water phase seeded with uranine is shown in green, while the dry pore and solid grain particles are dark. The fluorescence intensity directly reflects the water thickness. During evaporation, the pore with the largest radius and the least capillary resistance is first invaded by air. The DF meniscus recedes into the pore channel with a stable curvature. Some water is retained in the grain corners (CF), with a thickness gradually decreasing from the DF meniscus to the end of pore channel (see Movie S1). The water pressure gradient \( P_{w,DF} > P_{w,CF} \) drives water transport from DF toward the dry region through CF. The white arrows in Figures 8b–8d display CF extending longer during evaporation (as shown in Figure 6c). Similarly, the time sequence images in Figures 8e–8h show the same CF development in the pore channel of MM5-glass (experiment 3; 61°C), whereas the pore space invaded by air shows a light green film (TFF) instead of dark as in MM5-Si (see Movie S2).

Owing to the water pressure gradient \( P_{w,DF} > P_{w,CF} > P_{w,TFF} \), the disappearance order of “duct flow–corner flow–thick-film flow” was captured in the time sequence images in Figures 8i–8l (see Movie S3). Water-filled pores are not invaded by air layer by layer, because of the random pore sizes. Hence, bypass air invasion often occurs, and air invades the pore channel from both sides until a liquid bridge (LB) is left, connecting the neighboring grains. In this case, CF and TFF at both sides flow away from the liquid bridge...
(Figure 8j). DF dried first, leaving the last water as a liquid bridge (Figure 8j). In Figure 8k, TFF and weak CF are still visible after the liquid bridge disappears; CF and TFF must flow toward the left, where the open surface is located. The flow direction is implied by the thickness gradient of the CF and the TFF uranine intensity difference. In Figure 8l, the CF at the lower grain particle has already dried out, but a weak TFF can still be seen. Eventually, the pore channel dried out and became dark.

4.2. Spatial Patterns of Corner Flow and Thick-Film Flow

Figure 9a shows a typical spatial pattern of the CF region bounded by the evaporation (yellow) and percolation (cyan) fronts in MM5-Si during stage 2 ($t = 240$ min, 61°C). In Figure 9b (enlarged red window in Figure 9a), the completely dry region and water-saturated DF region can be recognized by the bright rings circling grain particles. These rings occur because light irradiating at the grain edges reflects back along the initial direction, which is the same for the isolated water clusters with small bright rings in the CF region (red arrow in Figure 9b). In partial wet pores, the CF slope eliminates this light artifact, presenting a dark edge. Figure 9c shows the same section as Figure 9b after image processing with a more straightforward segmentation; the DF region and isolated water clusters are marked in blue, the dry region is marked in yellow, and the partial wet pores are marked in green, with CF and liquid bridge marked in red. The isolated single-channel or multichannel water clusters force CF to flow along a highly tortuous path from the percolation front toward the evaporation front (blue-dashed line in Figure 9d), until the water clusters dry up and only a liquid bridge remains, the liquid bridge allows CF to pass from one grain particle to the neighboring particle.

Figure 10a shows a typical spatial pattern of the film region of MM5-glass during stage 2 ($t = 239$ min, 61°C). The film region, including both CF and TFF, is bounded by the evaporation (yellow) and percolation (cyan) fronts. The film region in MM5-glass displays a larger extension than the CF region in MM5-Si. In Figure 10b (magnified red window in Figure 10a), the completely dry region can be recognized by bluish pore channels, while the pore channel displays a brownish color when the inner surface is wet. As TFF flow
has a smaller curvature radius than CF, DF can supply a TFF path longer than the CF length. Therefore, CF close to the evaporation front (TFF front) is very weak and no longer connected with DF but is maintained in the form of a liquid ring surrounding the grain particles (Figure 10d). Liquid rings are indicated by the white circles in the original experiment image (Figure 10b) and marked in red in the processed image (Figure 10c). The liquid ring can be either isolated or connected with few neighboring grain particles by the liquid bridge; eventually, the liquid-ring water is transported from the corners toward TFF as evaporation supply.

The film region presents a whitish color caused by a light reflection (see Figure 10e). The processed image in Figure 10f shows that the corner flow near the percolation front is mostly connected with duct flow or isolated water clusters. With a sufficient water supply from the duct flow and isolated water clusters, the corner flow near the percolation front is thicker (Figure 10g) compared to the retained liquid ring near the evaporation front, which has no water supply (Figure 10d).

Table 1

| Detachment Time $t_c$, Constant Evaporation Rate $Q_{ev}$ of Stage 1, and the Relative Error Between the Gravimetrical and IP-Derived Mass Loss for All Experiments |
|---|---|---|---|---|---|---|---|
| $t_c$ [min] | $Q_{ev}$ [mg/min] | Relative error [%] |
| Exp.1 (RT) | Exp.2 (42°C) | Exp.3 (61°C) | Exp.1 (RT) | Exp.2 (42°C) | Exp.3 (61°C) | Exp.1 (RT) | Exp.2 (42°C) | Exp.3 (61°C) |
| MM5-Si | 40 | 12 | 6 | 0.7 | 2.5 | 6.4 | 1 | 11 | 3 |
| MM5-glass | 60 | 25 | 7 | 0.7 | 2.6 | 7.5 | 9 | 6 | 9 |
In summary, during the evaporation process, a pore channel may hold isolated water clusters after the neighboring pore channels are air invaded, but the water cluster is still hydraulically connected to the DF region through CF. When the isolated water cluster pore is also air invaded, the water cluster shrinks, and water is transported through CF (and TFF in MM5-glass) until only a liquid bridge is left. The liquid bridge connects the neighboring grain particles and allows the CF supply from the DF region or other isolated water clusters to continue. In MM5-Si, once a liquid bridge is broken, the residual CF becomes isolated as a liquid ring. The liquid ring dries out quickly without water supply, dragging the evaporation front away from the open surface. In MM5-glass, because of the water pressure gradient $P_{w,DF} > P_{w,CF} > P_{w,TFF}$, DF can supply a large TFF extension and maintain the evaporation front close to the open surface. After a liquid bridge is broken, the residual liquid ring serves as a water supply for TFF and further prevents the evaporation front from receding into the porous media.

4.3. Impact of Thick-Film Flow on Evaporation Rate

4.3.1. Mass Loss Curve

The water mass loss curves of MM5-Si and MM5-glass at the three temperatures are shown in Figure 11 with inserted experimental relative humidity and ambient temperature figures. The experimental data are renormalized as the ratio of the real mass loss to the micromodel's total water capacity. Meanwhile, experimental data given by the digital balance are smoothed by the “moving average” method (Hyndman, 2011) with a block size of 9. We tested the accuracy by comparing the mass loss data sets derived from digital balance and image processing (IP) for each experiment. The two data sets concurred, with relative errors between 1% and 11% for all experiments (Table 1).

As the kinetic energy of a molecule is proportional to its temperature, with an increase in temperature, the surface molecules tend to move rapidly until some molecules escape into the atmosphere. Therefore, the evaporation flux is enhanced by higher temperatures (Figures 11a–11c), and water flux is adapted to match...
the increased evaporation rate. The detachment time $t_c$ (end of stage 1) is defined when the averaged evaporation front recedes below the first layer of grains, so that no CF climbs up to the open surface along the grain edges. The duration of stage 1 ($t_c$) decreases with increasing temperature as the evaporation rate $Q_{ev}$ of stage 1 is larger in higher temperature and results in faster hydraulic detachment. The water transport in the micromodel cannot meet the significant demand of $Q_{ev}$ at high temperatures (42°C and 61°C), causing a dramatic transition in the mass loss curves between stages 1 and 2. Figure 11d shows an example of the linear fit of the mass loss curve for stage 1 (green line), and the slope determines the $Q_{ev}$ (stage 1). The $Q_{ev}$ of stage 2 can be derived from the slope of the time-dependent tangent line of the mass loss curve (Figure 12). Evaporation is dominated by water redistribution through capillary "pumping" in stage 1; therefore, the mass loss curves of the two micromodels overlap significantly in stage 1 (Figure 11).
Evaporation is enhanced by the TFF for two reasons. First, at the same temperature, after CF detaches from the open surface, TFF can still reach the open surface in MM5-glass, maintaining a constant evaporation rate \( \text{ev}_{\text{EQ}} \) and prolonging stage 1 (Figure 12). Meanwhile, MM5-Si enters stage 2 with a sharp decrease in the evaporation rate. The onsets of stage 2 occur at around 5-mm evaporation depth in MM5-Si, while the onsets occur at a later stage in MM5-glass, that is, at the evaporation depth around 7 mm for RT and 61°C and 10 mm for 42°C (Figures S1c, S2c in Supporting Information S1). Stage 1 was prolonged the most in the room temperature experiment, where the TFF transport was sufficient to compensate for the CF transport loss for evaporation.

The second reason is that in stage 2, the MM5-glass evaporation front is close to the open surface (before DF is dried out), while the evaporation front in MM5-Si continually recedes from the open surface. Further

Figure 11. Normalized time-dependent mass loss curves of MM5-Si and MM5-glass in (a) room temperature, (b) 42°C, and (c) 61°C. Mass loss data derived from digital balance measurements and image processing are indicated by colored and plus-symbol colored lines, respectively. The insets show the ambient temperature (a) and relative humidity, (a–c) during the experiments. (d) Stage 1-stage 2 transition of experiment 3 (61°C), where stage 1 and stage 2 are divided by a blue-dashed line, the equation describes the best linear-fit curve (green line) for stage 1.
explanations on evaporation front depth $x_p$ in both micromodels are discussed in the next section. With a certain boundary layer thickness $\delta$, the smaller $x_p$ in MM5-glass causes a larger vapor concentration gradient for diffusion and, consequently, a higher evaporation rate $Q_{ev}$ (Equation 3). Therefore, in stage 2, the $Q_{ev}$ of MM5-glass is always greater than that of MM5-Si, and the mass loss difference between the two micromodels increases with time. At 4 h, the mass loss was nearly doubled by the TFF in the 42°C and 61°C experiments.

Lower relative humidity can enhance evaporation through a higher vapor concentration gradient (Equation 3); this influence is insignificant in high temperature (experiments 2 and 3) but can be crucial in room temperature experiments. The two temperature curves in experiment 1 are similar, while the relative humidity (RH) curves diverge after 100 min (Figures 11a). We believe that the enlarging RH gap of up to 7% is sensitive enough to increase the $Q_{ev}$ in MM5-Si and compensates for the mass loss difference between the two micromodels in stage 2.

### 4.3.2. Geometric Characteristics of the Evaporation Process

The percolation front depth $x_p$ was derived by the geometric averaging of the front. The percolation front depth curve versus time (Figure 13b) show the same trend as the corresponding mass loss curve (Figure 11): TFF in MM5-glass enhances evaporation, resulting in a larger $x_p$ in MM5-glass than MM5-Si (at the same temperature).

In the pore network, a single pore channel is created by the grain walls and has a limited length based on the grain lengths. After DF in a pore channel vanishes, the water supply for longer CF is cut off from this pore channel, and CF is maintained by the DF from the adjacent or further pore channels by the liquid bridge. The liquid bridges near the evaporation front are vulnerable and can easily disappear owing to intensive evaporation. In MM5-Si, once the liquid bridge is broken, CF near the evaporation front loses hydraulic supply and will dry out instantly; thus, the evaporation front continues to recede along with the percolation front (Figure 13a), and the CF region is maintained with a relatively stable extension in stage 2 (Figure 13c). In MM5-glass, TFF spontaneously carpets the rough inner surface and is independent of the liquid bridges. Before the DF dries out, no restrictions, such as the absence of a liquid bridge, stop the TFF from growing. DF can maintain a long TFF, but only a limited CF extension; the liquid ring near the evaporation front is
isolated in stage 2 (Figures 10c and 10d), and TFF becomes the dominant water transport mechanism for evaporation. After an initial, short receding, the evaporation front in MM5-glass remains close to the open boundary due to the continuously extended TFF region, while the evaporation front in MM5-Si recedes continuously, resulting in a higher evaporation rate in MM5-glass than MM5-Si.

4.4. Viscous Length Scale Analysis

Physically, the extension of the CF and TFF regions during stage 2 is described by the time and temperature dependence of the viscous length

\[
L_{\text{visc}}(t, T) = k \cdot A \cdot \Delta p_w \int [\mu(T) \cdot Q_w(t, T)] dt.
\]
where \( k \) is the permeability \([\text{m}^2]\), \( A \) the cross section \([\text{m}^2]\), \( \mu \) the dynamic viscosity \([\text{Pa} \cdot \text{s}]\), \( \Delta p_{\text{w}} \) the driving water pressure difference \([\text{Pa}]\), and \( Q_\text{w} \) the internal water flow (CF or TFF) \([\text{m}^3/\text{s}]\). The internal water flow must satisfy the boundary condition at the evaporation front, that is, is controlled by the time \( t \) and temperature \( T \) dependence of the evaporation rate \( Q_\text{ev}(t,T) \). A steady state is maintained between water flow and evaporation, because the balancing diffusion process within the gas phase is always faster than the slow internal water flow. To discuss the \( T \) dependence, one has to discuss two competing processes. First, the significant decrease of the water viscosity, that is, \( \mu(60^\circ) \approx 0.5 \mu(\text{RT}) \), which causes an increasing \( \text{visc}_\text{EL} \). Second, the increase of the evaporation rate (higher diffusion coefficient), which causes a decreasing \( \text{visc}_\text{EL} \). Obviously, the viscosity effect dominates, as Figure 13c impressively demonstrates and as we have shown for CF (Geistlinger et al., 2019). Hence, \( \text{visc}_\text{EL} \) will increase with temperature.

We think that the inverse behavior of the CF region, the extension at which the higher temperature (42º; blue curve with crosses in Figure 13c) is smaller compared to that at lower temperature (RT; red curve with crosses), is caused by averaging the fractal evaporation front. The higher fractality of the evaporation front at 42º yields the expected higher mass loss (Figure S1a in Supporting Information). The fractality impact will be discussed in the next section.

Let us then discuss the time dependence. From Equation 5, it follows that \( L_\text{visc} \) will increase with time, because \( Q_\text{ev}(t,T) \) and hence \( Q_\text{w} \) will decrease with time (decreasing vapor concentration gradient). The \( t \) dependence of the viscous length \( (=w(t)) \), the mass loss, and the evaporation rate (green curves: \( \sqrt{t} \) behavior, red curves: linear \( t \) behavior) during stage 2. If viscous forces dominate the internal water flow, the viscous length and the mass loss are described by a diffusion-like \( \sqrt{t} \) behavior (Brutsaert & Chen, 1995). We observed this \( \sqrt{t} \) behavior both for soils (Geistlinger & Leuther, 2018); for CF in silicon micromodels (Geistlinger et al., 2019) and for TFF in glass-ceramic micromodels (Geistlinger et al., 2016). In the present study, this \( \sqrt{t} \)-behavior is observed with a high statistical significance (regression coefficients near 1) both for the mass loss (Figure 11; Figures S3a, S3b, S3d, and S3e in Supporting Information) and for the extension of the CF and TFF regions (Figure 13c; Figures S4a, S4b, S4d, and S4e in Supporting Information).
Although both micromodels exhibit a similar $\sqrt{t}$ behavior, the $t$-dependent increase of the TFF region is stronger compared to that of the CF region; e.g., the TFF region is 5 times larger than the CF region at 100 min for experiment 2 (42°) (compare the blue curves in Figure 13c and Figures S4b and S4e in Supporting Information S1), and the slope of the TFF curve is about 10 times larger. This indicates that the driving “force” (water pressure difference $\Delta \rho_w$ in Equation 5) is larger for TFF than that for CF. For CF, the water pressure difference is given by the curvature at the top and that at the root of the CF (see Figure 6). Taking into account that the critical contact angle within a rectangular geometry is 45° (Zulfíqar et al., 2020) and that the contact angle of the silicon micromodel is about 40° (Geistlinger et al., 2019), there is a weak curvature at the CF-top, which causes a weak driving “force.” Hence, the weak hydrophilic wettability of the smooth Si-surface is responsible for this weak driving “force.”

Naively, one would think that thick-film flow on a rough glass surface with an intrinsic contact angle of about 30° also causes a weak driving “force.” However, the wettability of rough surfaces is controlled by the degree of roughness, $r$ (=ratio of rough surface area to flat surface area; Wenzel, 1936; Levinson et al., 1988; Bico et al., 2001; Zulfíqar et al., 2020). Based on Wenzel’s argument, surface roughness amplifies hydrophilic wetting behavior. The driving force is given by a positive spreading parameter $S$ (Levinson et al., 1988). Physically, it means that the Free Energy of the 3-phase system is lowered, if the water film wets the rough surface (see Figure 14a). For a realistic roughness degree of 2 (cavity of regular triangle), one obtains a critical contact angle of 60° (Zulfíqar et al., 2020). Consequently, for all intrinsic contact angles smaller than this critical value, complete wetting or spontaneous thick-film flow occurs. The linear growth of TFF extension versus cumulative mass loss (mm) further proves the complete wetting and spontaneity (Figure S2d in Supporting Information S1). This qualitative change from partial wetting ($\theta = 30°$) to complete wetting by surface roughness causes the strong driving “force” of TFF.

Increasing the temperature up to 61° (experiment 3), we discover an interesting transition from $\sqrt{t}$ to linear $t$ behavior for both the mass loss and the extension of CF/TFF region (see Figure 11, Figures S3c, S3f in Supporting Information S1 and Figure 13c, Figures S4c, S4f in Supporting Information S1, respectively). This means a transition from $1/\sqrt{t}$ to constant evaporation rate (Figure 14) and that at late stage 2, the steady-state vapor-concentration gradient becomes very weak and tends to be constant.

Again, the strong increase of the TFF-region extension compared to that of the CF region is surprising. We believe that with an increasing temperature, evaporation from the large gas-water interface (=film surface; see Figures 14a and 14b) leads to a thinning effect. Hence, attractive long-range interaction becomes more dominant in thinner films, which allows the thick film to flow faster (linear $t$ behavior) and longer (larger TFF region). A rough estimate of the film thickness verifies this physical picture. We obtain for increasing temperature from RT to 61°, a decreasing film thickness from 34 to 13 µm (see Figure S5 in Supporting Information S1).

We note that the $m_{\text{loss}}$ data and the $w$ data are obtained by two independent measurements (=two IP-routines: (a) $m_{\text{loss}}$ is obtained by summing overall dry duct channels (region above the percolation front) neglecting the small retained water mass of TFF and (b) geometric evaluation of the evaporation and percolation front, that is, the CF/TFF region). Therefore, the $m_{\text{loss}}$ obtained by IP is always smaller than the gravitational $m_{\text{loss}}$ obtained by weighing.

### 4.5. Fractal Evaporation and Percolation Front

Evaporation has clear similarities to a drainage process, as is a fluid displaced by a lighter and less viscous fluid, and the pore invasion is determined by the pore size distribution. The probability of pore invasion is random in space because of the randomly distributed pore sizes; therefore, the evaporation pore invasion resembles invasion percolation, and the percolation and evaporation fronts hold the same fractal characteristic as a drainage front (Prat, 1995; Shaw, 1987; Yiotis et al., 2010). These expectations are corroborated by the highly fluctuating and fractal behavior of the percolation and evaporation fronts observed in experiment 3 at 240 min as illustrated in Figure 15.

Knowing the IP-derived $x_i(t)$ values (see Figure 13a), one can calculate the evaporation rate (Equation 3) and through integration, the theoretical mass loss (see Supporting Information S1). This mass loss (red
curve in Figure 16) is too small compared to the experimental mass loss (black curve in Figure 16). Taking into account the fractality of the evaporation front by the fractal gas-water interface, \( A_{\text{gw}}^{\text{fractal}} \) yields an excellent agreement for MM5-Si for higher temperatures 42°C and 61°C (blue curves in Figure S6b in Supporting Information S1 and Figure 16). At RT, the fractal curve overestimates evaporation (Figure S6a in Supporting Information S1). The reason for the RT deviation is not clear. We suppose that IP thresholding gives too much empty menisci, where they still have some water inside, and the evaporation front is smoother (smaller) than after IP. This would explain why the fractal IP curve overestimates evaporation.

Contrary to the real physical behavior of the evaporation front in a porous medium, the SSC model assumes that the evaporation process can be described by an effective single-square capillary, that is, by a flat averaged evaporation front (see Figure 2, definition of the averaged fronts). This causes a fundamental inconsistency between the time dependence of the mass loss and that of the x position of the flat evaporation front (see Geistlinger et al., 2019).

5. Summary and Conclusions

This study investigates the impact of thick-film flow (caused by surface roughness) on the evaporation process. We conducted a series of evaporation experiments at different temperatures (room temperature, 42°C, and 61°C) with two micromodels (80 × 80 quadratic lattice pore structure with lognormal pore size distribution). The MM5-Si and MM5-glass micromodels were produced by silicon and glass ceramics, respectively, to represent the smooth (MM5-Si) and rough (MM5-glass) inner surfaces. Hence, thick-film flow only occurs in MM5-glass and corner flow occurs in both MM5-Si and glass. MM5-glass was constructed by an anisotropic photolithographic etching process based on the photosensitive glass-ceramic FOTURAN. MM5-Si was constructed using the interval-based ICP-DRIE technology, guaranteeing the high edge steepness (sharp corners) and the true mapping of the lattice structure.

Since both micromodels possess an identical pore structure, they show a similar stage1-behavior, which is controlled by water redistribution.
through capillary "pumping." If the menisci have receded into the pores, water flow still exists within the pore corners and along the rough pore surfaces. Stage 2 is controlled by the internal water flow and the different driving forces of corner flow and thick-film flow are responsible for the different stage 2 behavior of the evaporation process. Our experimental study shows that the interplay of surface roughness and wettability plays a key role for the time and temperature behavior of the evaporation process. We found a similar $\sqrt{t}$ behavior up to 42° and a transition to linear $t$ behavior for higher temperatures (61°) with a high statistical significance (regression coefficients near 1) for both micromodels. Our experimental results elucidate the strong temporal correlation between mass loss and geometric pattern of the unsaturated CF and TFF regions. Both show a similar time behavior with a high statistical significance. However, the glass-ceramic micromodels always exhibit a stronger increase of the evaporation characteristics (mass loss and extension of the CF/TFF region) with increasing time and temperature. The weak hydrophilic wettability (contact angle of approximately 40°; partial wetting) of the oxidized silicon micromodels causes a weak driving force for the corner flow. This results in less mass loss and smaller viscous flow length (=extension of the CF region). In contrast, surface roughness of the glass-ceramic micromodel causes complete wetting and hence a strong driving force, which results in higher mass loss and larger viscous length compared to those of the Si micromodel.

Based on the difference between IP-derived mass loss and gravitational mass loss, we estimated the film thickness of the TFF and found an interesting film thinning effect with increasing temperature. The film thickness decreases from 34 to 13 µm if the temperature increases from RT to 61°. We attribute this effect to a stronger evaporation caused by the large gas-water interface of the thick-film flow.

For a consistent description of the time-dependent mass loss and the geometry of the CF/TFF region, one has to take into account the fractality of the evaporation front. An average flat evaporation front always leads to a principal inconsistency, which is inherent to the SSC model as we have shown in Geistlinger et al. (2019).

This evaporation study was based on an artificial regular pore structure with an ideal rectangular channel geometry. We will continue our evaporation study with an irregular pore structure, which was derived by a new 3D-2D mapping method (Zulfiqar et al., 2020) from 3D-micro-CT image of real soil. Therefore, the microstructure exhibits the similar morphological, topological, and geometrical properties as the soil structure.

Data Availability Statement

The data can be accessed at this site (https://www.ufz.de/index.php?en=39791).

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