

Shell and lumen side flow and pressure communication during permeation and filtration in a multibore polymer membrane module

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Abstract

This study reveals the importance of the module geometry on the flow field and pressure distribution during membrane permeation for multibore membranes. The pathways of permeation are unraveled within a custom-made single multibore membrane module. For this, we combine flow velocimetry of magnetic resonance imaging (MRI) with computational fluid dynamic (CFD) simulations and permeation experiments. First, a systematic simulation study identifies flow patterns based on simplified geometry features that are supported experimentally through flow-MRI measurements. This comprehensive study shows how small geometric deviations from the idealistic assumptions result in unexpected fluid flow on the shell and lumen side in the module. Second, the influence of those non-ideal flow patterns during the filtration of silica particles are revealed by MRI. The results indicate heterogeneous silica deposition due to geometry induced flow fields. Contrary to the idealized assumption, the subsequent backwashing is also influenced by those deposition patterns. Hence, unavoidable non-idealities of membrane positioning during the construction of the module influence the performance of the membrane filtration. With this study, we stimulate to analyze and pioneer new strategies to fully recover the membrane's performance after filtration cycles during backwashing based on a precisely designed backwash optimized permeate channel geometry in membrane modules.

Keywords: Multibore (multichannel) membrane, Computational fluid dynamics (CFD), Flow magnetic resonance imaging (MRI), Fouling, Backwashing

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1. Introduction

Multibore membranes are a collection of various single bore hollow fiber membranes combined in a multibore geometry resulting in significant robustness improvement. [1–4]. Due to their superior tensile properties, multibore membranes have found their way into many applications such as drinking water production and seawater desalination [1, 2, 5], and wastewater and surface water filtration (direct contact membrane distillation [3, 6, 7] and vacuum membrane distillation [8–10]).

This wide variety of applications led to several studies optimizing their geometry [2, 3, 7, 11–13] and the membrane material [8, 13, 14]. Generally, the bore channels are straight, yet in novel geometries, the multibore membranes can be fabricated having a spiraling twist through the use of 3D-printed rotating spinnerets [15]. These twisted tribore hollow fibers redirect the flow direction and thus impair concentration polarization effects.

Multibore membranes have been introduced as polymeric membranes made of polyether-sulfone (PES) [1, 3, 5, 15, 16] as well as polyvinylidene fluoride (PVDF) [6–10] with high water permeability and high pH tolerances. Ceramic multibore membranes are used for harsher environments [17–21].

Long-term filtration performance strongly depends on the membrane’s backwash efficiency. After each filtration cycle, a cake layer has been built on the membrane surface and needs to be removed to recover the intrinsic membrane permeability. This is accomplished by a backwash cycle during which the fluid flow is reversed in direction, and the cake layer is being pushed off the membrane surface and out of the membrane lumen. The relatively large outer diameter of a multibore structure results in a lower pressure loss in a multibore bundle allowing a more even water distribution during a backwash cycle and thus improving its efficiency [1, 2]. In this way, the use of chemical agents for cleaning such as chlorinated water may be reduced [5].

However, fundamental knowledge of internal flux pathways and particle deposition is still subject to current research in multibore membranes [20]. The recovered permeability typically evaluates the effect of backwashing [5, 22]. A novel methodology to evaluate

the membranes filtration performance and backwashing capabilities is magnetic resonance imaging (MRI). MRI and flow-MRI are non-invasive tools to monitor fluid velocities and particle depositions online and in-situ [15, 23–29].

An innovative approach to extending the measurement capabilities for porous multibore membranes by MRI and flow-MRI are validated computational fluid dynamics (CFD) simulations [15]. So far, Kagramanov et al. and Frederic et al. simulated fluid flow in ceramic multibore membranes with homogeneous pore size distributions [30, 31]. The majority of studies thereby focus on optimizing the membrane itself. However, shell sided hydrodynamic effects in hollow fiber membrane modules have an equally pronounced impact on the membrane’s performance. The tangential inlet and outlet flow influence the flow field inside membrane modules as validated by CFD simulations and particle image velocimetry (PIV) [32].

Recently, Schuhmann et al. used MRI to investigate the filtration behavior in multibore membranes [33]. The authors hypothesize that the location of the membrane in the module might influence the velocity field. The manuscript also provides some first insight through inside-out dead-end filtration and at constant feed pressure at a single position only. While the study is a good first approach towards understanding the hydrodynamic and filtration phenomena in multibore membranes, it has the inherent disadvantage of a constant pressure experiment where the flux and hence the local velocities continuously vary over time.

In this study, we investigate the hydrodynamics inside a custom made multibore membrane module inspired by the work of Hawthorne in 1951 [34] on secondary flows within curved geometries. The positioning of the membrane inside the module affects the shell and lumen side flow and pressure communication during membrane operation. We combine the capabilities of MRI, flow-MRI, CFD simulations, and permeation experiments to unravel the pathways of flux during filtration and backwash fully. CFD simulations are used to interpret the complex velocity fields. The richness of information of the flow is visualized in multiple cross-sectional planes in vector plots for radial velocities and velocity magnitude plots for axial velocities. Next, we analyze silica particle deposition at different positions within the membrane in dead-end as well as cross-flow mode by MRI and flow-MRI. Here, we chose

constant feed flux to isolate the effects of directed flow velocity on fouling. With this study, we evaluate the efficiency of backwashing in multibore membranes pioneering the pathway to find strategies to recover the membrane's permeability fully.

2. Materials and Methods

2.1. Membrane

Multibore membranes used in this work are commercially available PES SevenBore™ fibers from SUEZ Water Technologies & Solutions [35]. They are commonly used in inside-out ultrafiltration operation. The membranes' internal structure features one center bore that is symmetrically surrounded by six bores close to the membrane's outside surface. SEM images of the membrane's cross-section are taken by a Tabletop Microscope TM3030 and by a Hitachi High-Technologies Corporation. To maintain the pore structure, the membrane is broken in liquid nitrogen. The cross-section of the membrane is displayed in different magnifications in Figure 1 i). Membranes' volume porosity is averaged over multiple slices and displayed in Table 1. The software ImageJ is used to determine the porosity by the area fraction measurement based on SEM images. Pure water flux measurements evaluate the permeability for the membrane module arrangement. Subsequently, the intrinsic permeability is calculated using Darcy's law.

	Unit	Value	
Bore diameter	[mm]	0.9	measured
Outer diameter	[mm]	4	measured
Porosity	[–]	0.82372	calculated
Intrinsic permeability	[m ²]	$1.1387 \cdot 10^{-15}$	calculated
Pure water permeability	[LMH bar ⁻¹]	1000	measured

Table 1: Measured and calculated physical properties of the polymeric PES SevenBore™ fibers from SUEZ [35].

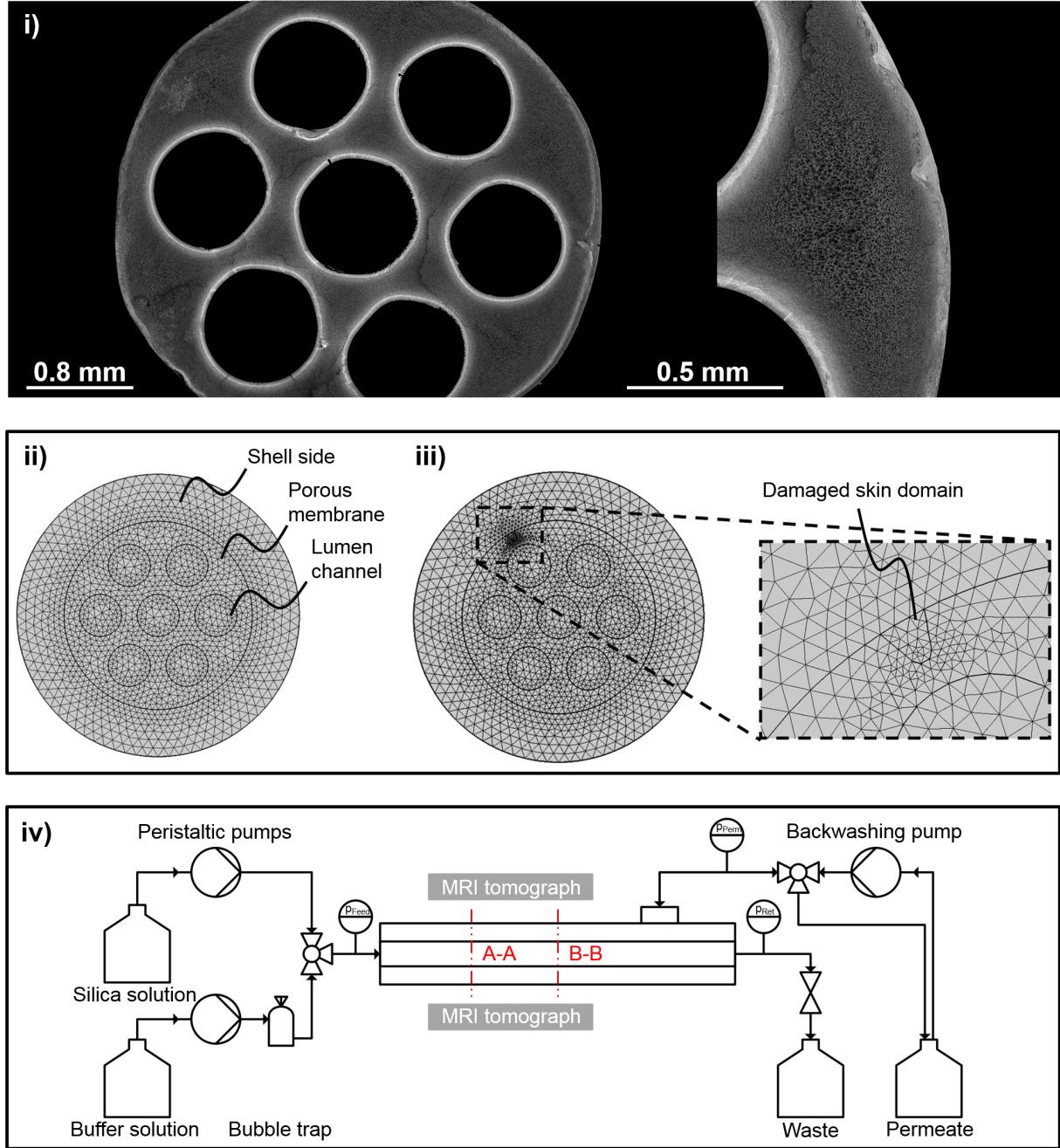


Figure 1: Overview of: i) Scanning electron microscopy (SEM) images of two cross-sections at different magnification of a SevenBore™ fiber from SUEZ [35]. The background of the SEM images is adapted for better visibility. Original images can be found in the supplementary material. ii) Mashed simulation domain of the lumen channels, the shell channel, and the porous membrane. iii) Additional simulation domain for the damaged outer skin of the membrane. iv) MRI set-up indicating the location of the measured slices (A-A and B-B) in the module.

2.2. Computational fluid dynamics (CFD) simulation

CFD simulations of the multibore membranes are performed using COMSOL Multiphysics[®] [36]. Throughout this study, pure water velocity fields are investigated by different geometry modifications. All investigated membrane module geometries are compared to the ideal reference geometry stated in Section 3.1. The numerical Finite Element Method (FEM) simulations solve the Navier-Stokes equations. All performed CFD studies use the *Free and Porous Media Flow* physics interface that combines the Laminar Flow interface and the Brinkman Equation interface. The open spaces of the flow channels (lumen and shell) are modeled as *free water flow*, and the membrane is considered as a homogeneous porous medium.

Boundaries of the simulation domains are defined by the 2-end module arrangement as shown in Figure 1 ii). The flow scheme is dead-end inside-out. Inlet boundaries are set as standard inflow velocity. The outlet boundary is a pressure outlet with suppressed backflow set to ambient pressure. The simulation utilizes water, for which the material properties are taken from the COMSOL library. The membrane is configured as porous media with user-defined porosity and intrinsic permeability. The material's density and dynamic viscosity correlate to the temperature of 293.15 K. Other physical properties of the membrane can be taken from Table 1. The respective mesh is optimized for each geometry study.

All simulations are implemented as a time-independent study. The parallel sparse direct solver MUMPS is used as a solver for the stationary fluid dynamic study.

By solving the Navier-Stokes equation by FEM, all information about the velocities of all spatial directions of a Cartesian coordinate system is retrievable for each node of the meshed grid. However, for comparability with the flow-MRI, the displayed infinitesimal 2D CFD slice plots are averaged over the same slice thickness of 1 cm as in the 2D flow-MRI. This procedure is performed as described in our previous work [15, 23]. The axial velocity magnitudes are average as $|u_{axial}| = \sqrt{u_z^2}$ and the radial velocity magnitudes as $|u_{radial}| = \sqrt{u_x^2 + u_y^2}$, respectively. Arrows in our figures depict the direction of flow.

2.3. MRI, flow-MRI measurements, and data processing

Magnetic resonance imaging is used as a non-invasive tool to visualize and evaluate the hydrodynamic conditions as well as the fouling behavior of the membrane inside the module. Within the tomograph, all proton spins align along the static magnetic field. During measurements, an oscillating magnetic field flips the spins out of their equilibrium state. After the dynamic magnetic field is shut off, the spins relax to their original position. The emission is captured as a signal. A subsequent Fourier transformation converts the decaying signal into an image. Overlaying flow and no flow images facilitate the capture of fluid velocities. This measuring technique using the 2D spin-echo pulse sequence is adapted from previous studies [15, 23, 28, 29, 37].

All MRI measurements are performed on a Magritek low-field NMR tomograph system. The tomograph operates at a Larmor frequency of 23.8 MHz with a field strength of 0.56 T. The membrane module is placed horizontally within the 60 mm inner diameter imaging probe (or birdcage). A bubble trap prevents air from entering the membrane and disturbing the signal of the tomograph. MRI imaging parameters are listed in Table 2. All images have a signal to noise ratio (SNR) of larger than 10. Measurement data from Magritek’s software PROSPA is post-processed using MATLAB to derive corresponding images of the axial and radial velocities [15, 28, 29].

Slices A-A and B-B (Figure 1 iv) have a distance of 3 cm. Subsequently, these MRI measurements are compared to simulations in terms of radial and axial flow distribution in a dead-end inside-out mode. We visualize the lumen side, transport paths inside the membrane, and the development of flow in the shell side. Additionally, backwash flows and deposited particles in dead-end and cross-flow mode are investigated by MRI and flow-MRI.

	Unit	Value
Repetition time	[ms]	600
Echo time	[ms]	35
Spatial resolution [x · y · z]	[mm · mm · mm]	[12 · 12 · 10]
Number of pixels [x · y]	[–]	128 · 128
Number of scans	[–]	12

Table 2: MRI parameter settings for the flow measurements of the multibore membrane module.

2.4. Filtration experiments - silica fouling and backwashing procedure

All filtration experiments are performed in a custom made membrane module as displayed in Figures 8, 9 and 11. Design and construction of the module are adapted from our previous study [15]. All connectors and junctions are additive manufactured with an Objet Eden 260V three-dimensional printing system from Stratasys. The module itself contains one multibore membrane. Flow modes inside the membrane module are adjusted with inlet and outlet valve control.

During filtration and backwashing experiments, the module is inserted in the MRI tomograph's bird cage for in-situ measurements (Figure 1 iv). All experiments are performed with a buffer solution of de-ionized water and 1.48 g L^{-1} copper(II) sulfate pentahydrate ($\text{Cu}[\text{SO}_4] \cdot 5\text{H}_2\text{O}$). The latter is used to reduce the T_1 relaxation time and therefore the overall measuring time. For dead-end measurements, the feed volume flow was set to $175 \text{ }\mu\text{L s}^{-1}$.

Fouling experiments are performed with an aqueous model solution of 0.3 wt% spherical silica particles [28]. The silica particles SYLOID C 803 (SiO_2) with a particle size of $3.4 \text{ }\mu\text{m}$ to $4.0 \text{ }\mu\text{m}$ are purchased from GRACE [38]. Dead-end silica fouling experiments are performed with a buffer solution volume flow of $125 \text{ }\mu\text{L s}^{-1}$ and silica suspension flow of $50 \text{ }\mu\text{L s}^{-1}$. We stopped our filtration experiment when the feed pressure reached 2.1 bar. Two backwashing cycles are performed after particle deposition experiments. For 30 min, a flux of 180 LMH and 360 LMH is applied, respectively. The feed valve remained closed during both backwashing steps. After each backwashing step, MRI and flow-MRI measurements were conducted to check whether or not the membrane was cleaned. Cross-flow experiments are performed with a volume flow of $350 \text{ }\mu\text{L s}^{-1}$ consisting of $200 \text{ }\mu\text{L s}^{-1}$ buffer solution and $150 \text{ }\mu\text{L s}^{-1}$ silica solution with a constant flux of 100 LMH.

3. Results

3.1. CFD velocimetry - ideal membrane position

First, we present the results of the reference module to evaluate the different non-ideal membrane positions. The membrane is positioned in the center of the module. Pure water flow enters the multibore membrane equally distributed in the seven bore channels and permeates in a dead-end arrangement through the porous structure to the shell side. Figure 2 displays the axial and radial velocity components during inside-out permeation at the center of the module (B-B). The evolution of the flow field in axial direction (A-A and C-C) can be found in the supplementary material (Supplement 1).

Due to a dead-end inside-out flow arrangement, the axial velocity magnitude in the seven bore channels decreases in axial direction whereas the axial velocity in the shell side increases with the flow through the module to maintain the mass balance. In the bore and shell channel, the flow profiles show a laminar parabolic flow formation. The shown velocity magnitudes between shell and bore channels differ because the cross-section of the shell is larger than the cross-section of the seven channels.

Arrows in Figure 2 depict the direction of the radial velocities, and their length corresponds to the velocity magnitude. The radial velocity magnitude in the channels decreases with the flow along the module in all seven bores. The radial velocity magnitude and the axial velocity magnitude correlate appropriately to the mass balance. Here, it is observable that the center bore already shows lower radial velocities close to the feed inlet. The arrows reveal that the flow is directed from the center to the shell side without diverting and follow the path of least resistance. The magnitude of the six outer bores differs from the center bore. Flow from the inner bore permeates over the entire circumference, through the membrane structure into the surrounding outer bore channels (this model assumes a homogeneous porous structure excluding higher resistance skin layers at the lumen side), while the water from the outer bores permeates straight in the direction of the shell. Here the flow is not distributed over the whole circumference. The results indicate that the structure of the porous membrane plays a crucial role in flow field development and demands a more

detailed investigation.

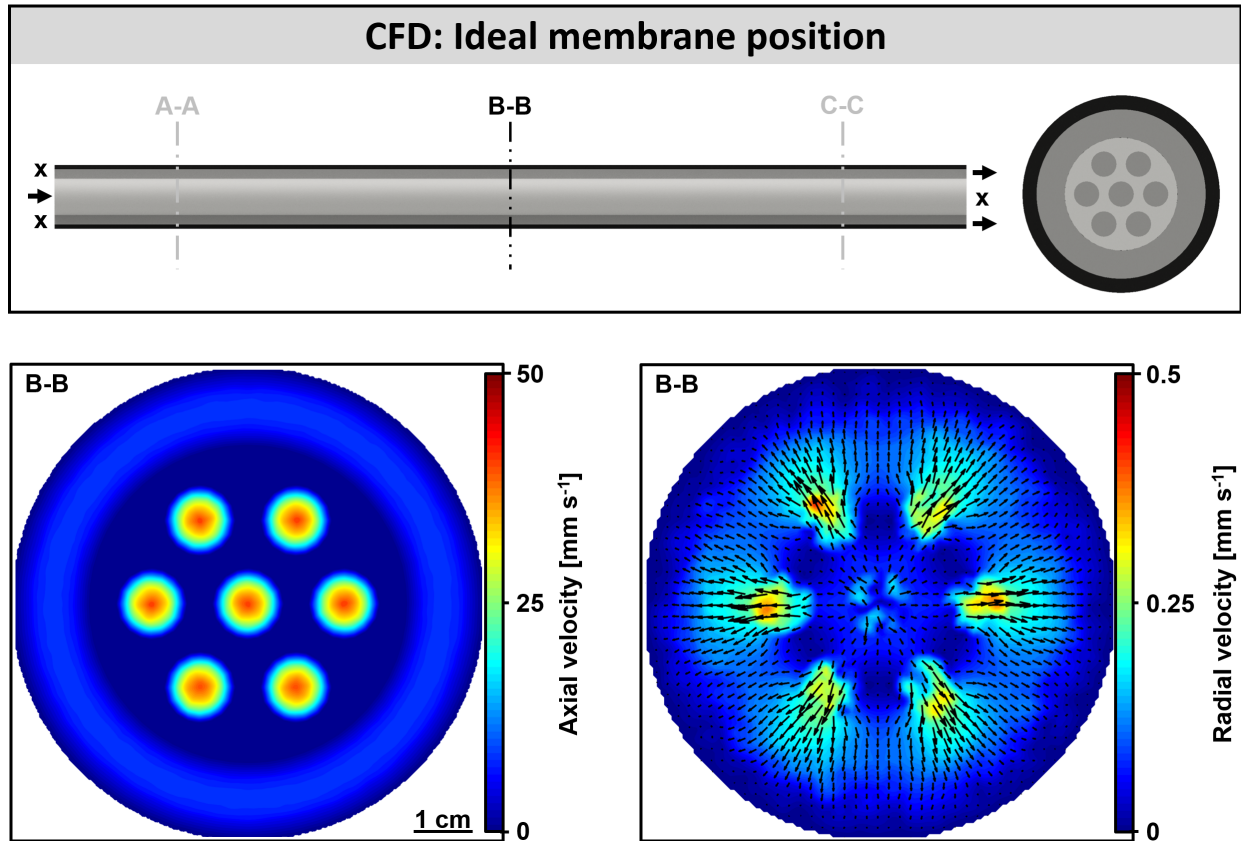


Figure 2: CFD velocimetry - ideal membrane behavior: Shown is the computed ideal membrane behavior during membrane inside-out permeation. No shell sided secondary flow field is indicated. The lumen flow is on a direct path to the shell side without diverted direction.

3.2. CFD velocimetry - excentric membrane position in the module

Next, our CFD study investigates the effects on the flow field if a membrane is positioned excentrically as this could easily be found in commercial modules containing a couple thousand of these multibore membranes. The results of the excentric membrane position are displayed in Figure 3. Similar effects as discussed in Section 3.1 can be observed. Opposite to the reference geometry, a radial flow field on the shell side is now induced. The fluid is forced from the minimal distance between the membrane and inner tube wall to the maximal distance. This effect results from a lower pressure drop of the annular gap flow with increasing gap distance (cf. Supplement 2). The excentric position does not influence the lumen to shell side directed flow through the porous membrane. This can be seen by the directed outward flow inside the lumen channel. Results of other slice positions (A-A and C-C) of the excentrically positioned membrane do not differ and can be found in the supplementary material.

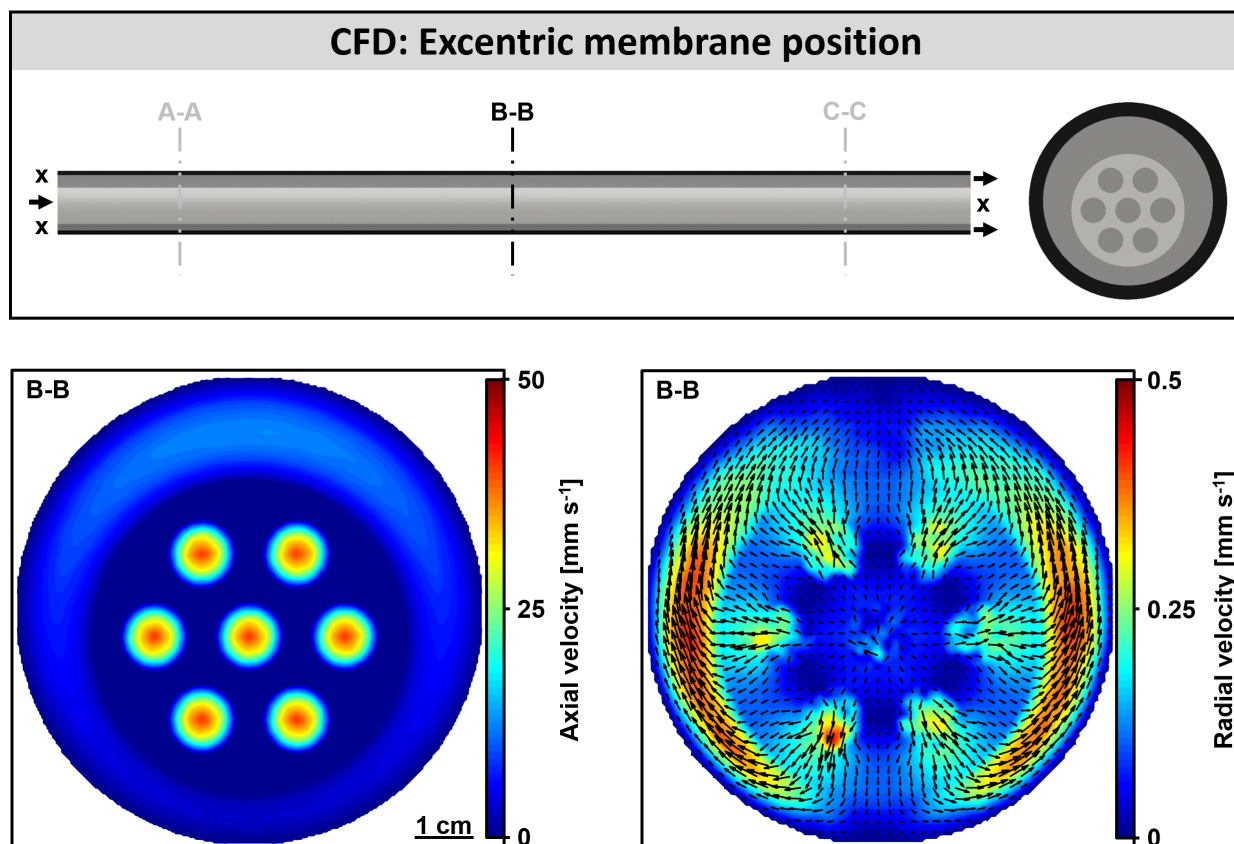


Figure 3: CFD velocimetry - excentric membrane position in the module: Shown is the middle slice of the computed membrane behavior during membrane inside-out permeation. The lumen flow is on a direct path to the shell side without diverted direction. A Shell sided secondary annular gap flow field is introduced due to the different distances to the tube wall.

3.3. CFD velocimetry - sagging membrane in the module

The hollow fibers bend during permeation because of their swelling behavior in contact with pure water. This sagging effect and its impact on the flow field is shown in Figure 4. The displayed slices correspond to the entrance (A-A) and the outflow (C-C) of the module. In B-B, the sagged membrane is in maximal deflection. Pressure distribution and flow fields for slices A-A, B-B and C-C are depicted in the supplementary information (Supplement 3).

Sagging of the membrane introduces a secondary flow in the lumen channel [34]. Oppositely, the radial velocity components are not only directed outwards during inside-out permeation. The sagging of the hollow fiber membrane introduces a radial directed flow towards the bending direction. The radial velocity magnitude is most strongly pronounced in two lumen channels at the outermost arc of the bending. The direction of the secondary flow is reversed after crossing the center point of the module. Now, the radial velocity magnitude is most strongly pronounced in the two opposite lumen channels.

Additionally, a shell sided secondary annular gap flow field is observed corresponding to the distance of the membrane to the inner wall of the tube wall. The curvature of the membrane results in an increased annular gap due to the different distances to the tube wall. The fluid is forced from the minimum distance between the membrane and inner tube wall to the maximum distance as observed in Section 3.2. This effect is reversed after crossing the inflection point at B-B.

The results of the sagged membrane suggest that the positioning of the membrane within the module causes a radial flow field formation. This effect is positively validated by the MRI velocity in Section 3.6. Flow conditions along and through the membrane influence the retention behavior and the resulting fouling behavior during operation. Hence, the sagging of the membrane causes the membrane to behave non-ideal. This kind of heterogeneous flow profile should be anticipated in commercial membrane filtration modules where perfect (multibore) fiber alignment typically does not exist.

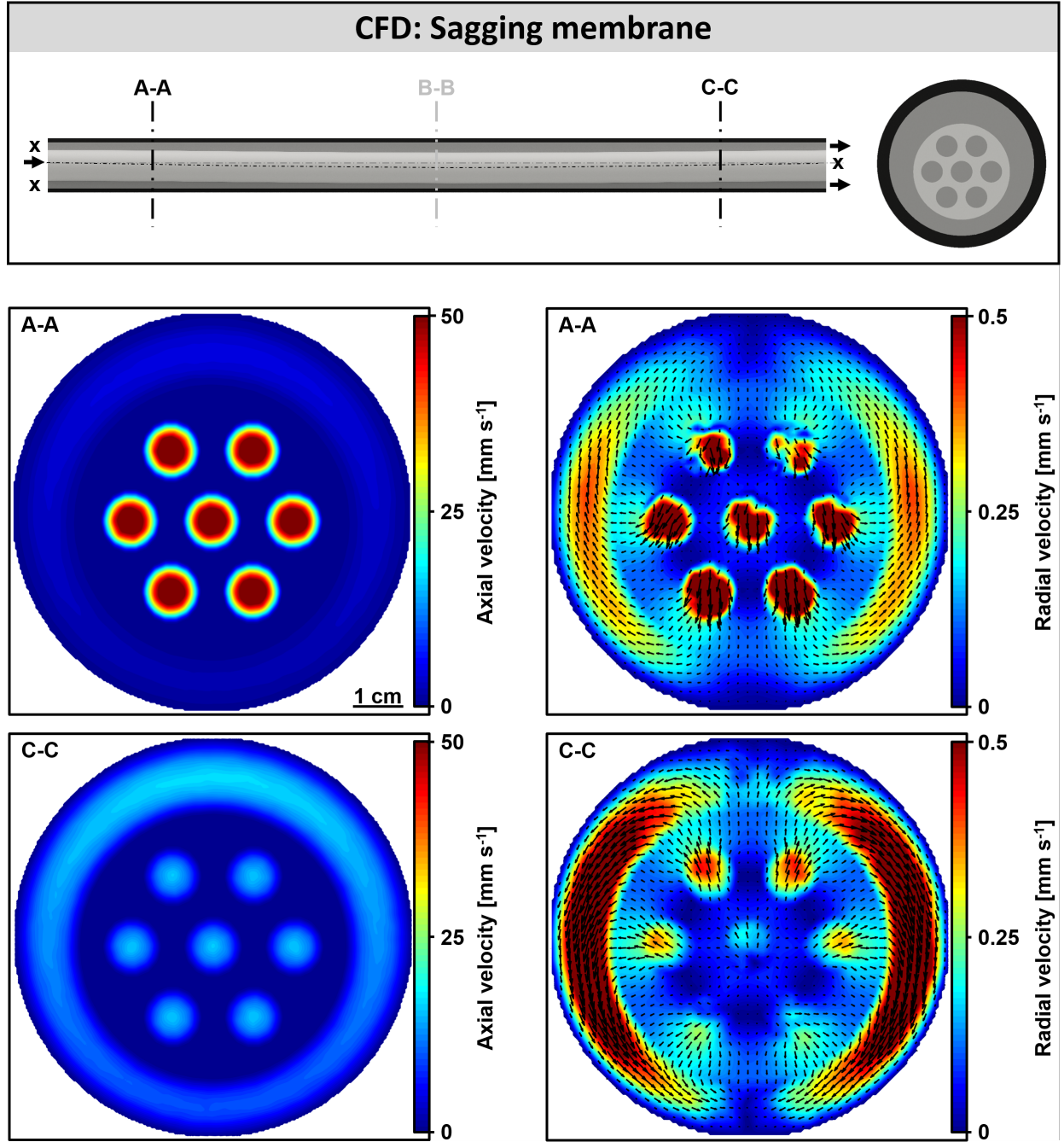


Figure 4: CFD velocimetry - sagging membrane in the module: Shown are the slices at the entrance and the outflow of the module. The slices correspond to the computed membrane behavior during membrane inside-out permeation. Sagging of the membrane introduces secondary flow fields in the lumen channel. A shell sided secondary annular gap flow field is observed corresponding to the distance of the membrane to the inner wall of the tube wall.

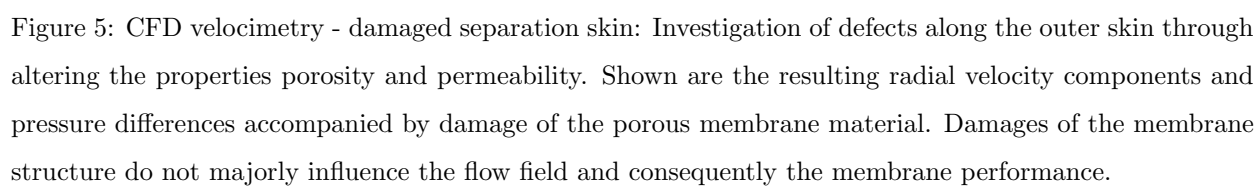
3.4. CFD velocimetry - damaged separation skin membrane

Membrane spacers are predominantly used in flat sheet spiral-wound modules and stacked flat sheet modules. These spacers define the channel geometry and act as mixing devices to reduce concentration polarization by redirecting the flow field. Equipping membrane modules with spacers may cause damaging of the membrane skin during module construction. Defects in hollow fiber membranes along the outer skin can occur during the fiber spinning process and fiber handling during processing into modules.

In the following, the impact of axial damages in the outside skin of the membrane is investigated. Altering the properties of porosity and permeability along the outer side of the fiber mimics this axial damage. The results of the corresponding CFD simulation are shown in Figure 5 (Supplement 4 and 5). The module is operated in dead-end inside-out filtration mode. The damaged domain can either have a higher or a lower intrinsic permeability. Due to the symmetry of the damaged domain only slice B-B is presented.

The damaged outer skin has no significant effect on the permeation performance of the membrane. The relative pressure illustration depicts the differences between higher and lower intrinsic permeability, respectively. A damaged outer membrane skin with higher permeability results in a higher flow from the lumen channel into the shell side. Damages that lead to a lower permeability show exactly the opposite effect. Due to the relatively high resistance of the porous membrane material, the surrounding lumen channels are not affected by a damaged outer skin. No bypass flow can be observed.

The separation layer of the multibore membrane is located at the lumen side. Therefore, damages at the membrane outside structure do not have a significant negative influence on the performance of the membranes. A benefit of inside-out hollow fiber membranes is that the skin is well protected.



3.5. CFD velocimetry - module flow field

Up until now, we assumed a standard velocity inlet to the seven bores. In the following, different flow entrance behaviors are investigated for the bore channels as well as a different outlet channel for the shell side of the membrane module.

3.5.1. Module flow field - the lumen sided flow

In Figure 6 a comparison is shown between the ideal reference module i) and an altered inlet flow ii). Additionally, Supplement 6 illustrates slices A-A and C-C. The altered inlet flow is based on the module used for MRI measurements. Here, the parabolic flow of the cylindrical domain enters the bore channels in front of the multibore membrane. As compared to the ideal reference module, the assumption that the volume flow is equally distributed into all bore channels does not hold. The inner bore channel does not take part in the filtration shown by the slice in B-B of the module. The volume flow seeks the path of least resistance. In the reference module, the pressure is the highest in the central bore and lowest in the shell domain. There is a pressure difference from the central bore to the outer bore and a pressure difference from the outer bore to the shell domain. The pressure difference corresponds to the driving force of the fluid. The CFD study assumes a homogeneous porous polymer matrix of the membrane domain. Therefore, the resistance from the inner bore to the shell side is higher in comparison to the resistance from the six outer bores to the shell side.

However, the SEM images in Figure 1 i) of the cross-section of the multibore membrane reveal a gradient in pore size. The solvent exchange process during membrane fabrication influences the formation of the separation skin and micro- to macro-voids. The separation skin forms a diffusive barrier which slows down the phase inversion process beyond the skin later creating asymmetry in the final polymeric structure. For ceramic membranes the assumption of a homogeneous pore size holds. Only the separation skin varies in ceramic membranes.

Flow-MRI results (Figure 8) validate the CFD study. The simulation and measurement results are in good agreement and suggest that the assumption of equal distribution of the

flow to the seven channels is a valid approximation of the measured membrane behavior. Future investigations should focus on modeling the local porosity and local permeability of the porous membrane matrix.

3.5.2. Module flow field - the shell sided flow

The module for MRI measurements has no colinear inlet and outlet in the direction of the membrane. The connection between the shell of the module and the ambient is perpendicular to the membrane direction. This flow arrangement is shown in Figure 7 (and in Supplement 7).

The perpendicular arrangement of the outlet in inside-out dead-end filtration does not affect the fluid flow as indicated by slice B-B. The outlet channel has no distinct influence on the lumen to shell sided permeation throughout the multibore membrane. Distinct characteristics as shown for the reference module in Section 3.1 are dominant. Only the flow is directed to the outlet in section C-C to maintain the mass balance in the dead-end arrangement.

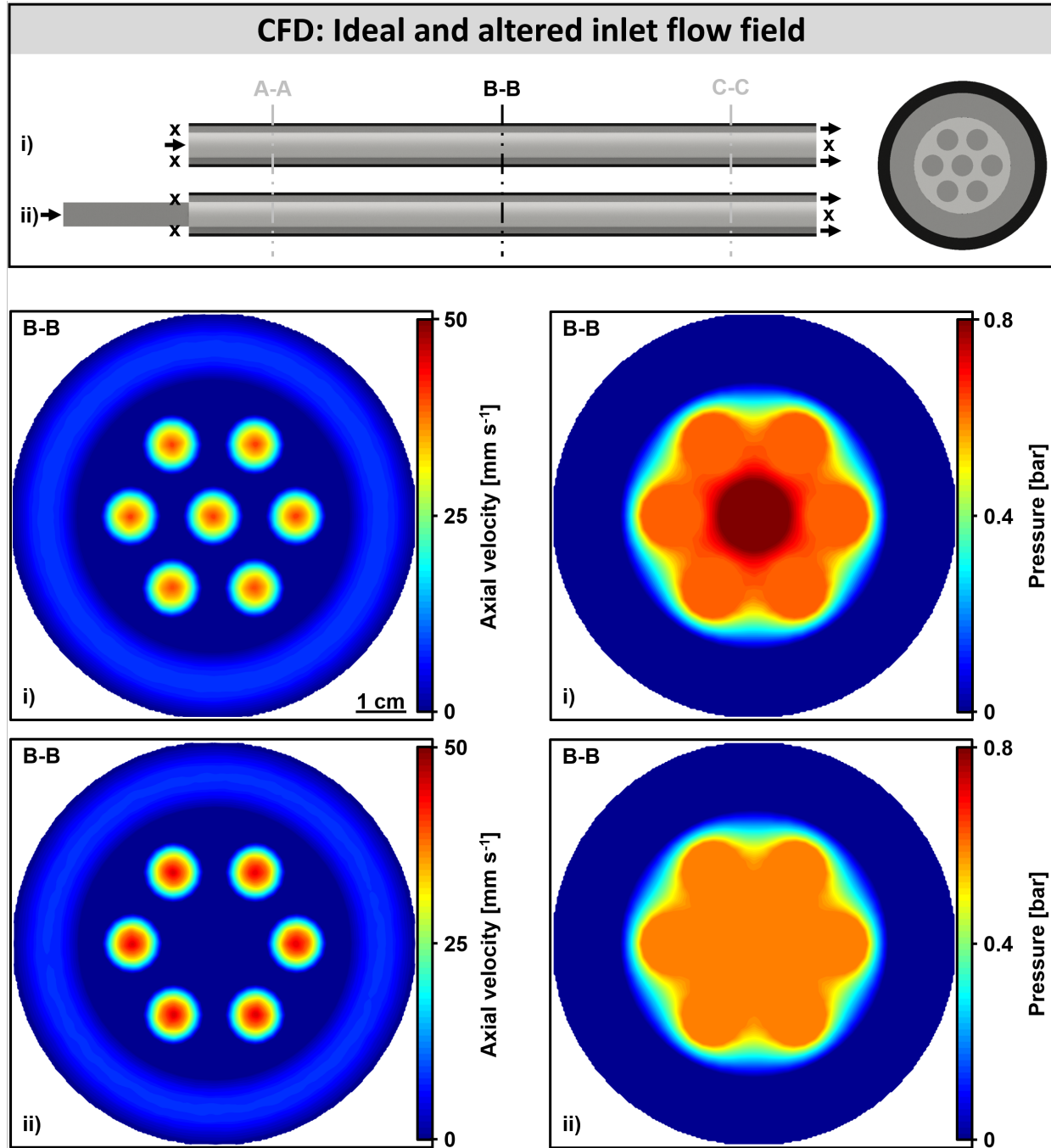


Figure 6: CFD velocimetry lumen sided flow field behavior: Shown is the investigation of different lumen sided inlet flow fields. i) The ideal reference module assumes that the volume flow is equally distributed into all lumen channels. ii) Parabolic flow of the cylindrical domain enters the lumen channels in front of the multibore membrane as suggested by the experimental module set-up. The comparison indicates that the entrance behavior of the fluid flow differs from experimental results. Hence, modeling the local porosity and local permeability gradients of the porous membrane matrix should be considered.

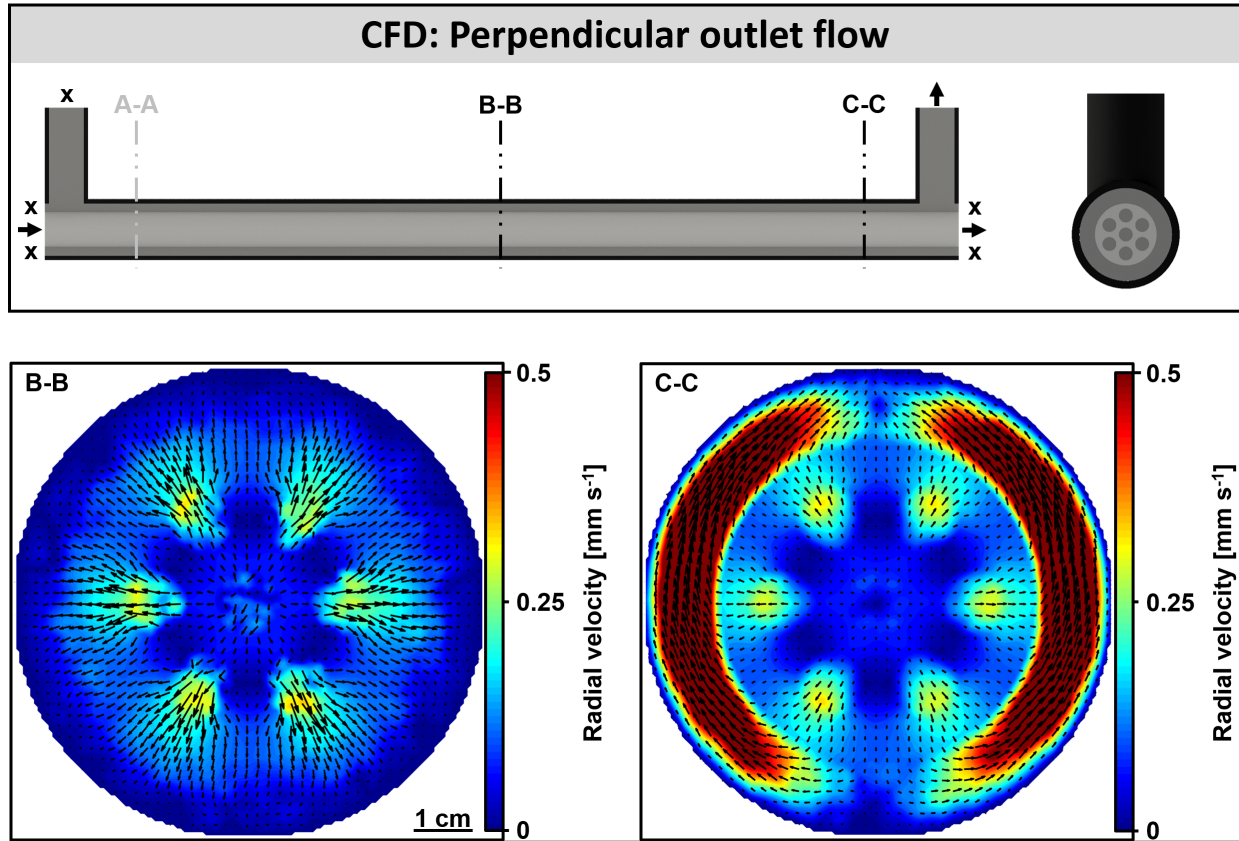


Figure 7: CFD velocimetry outlet sided flow field behavior: Shown is the effect of the fluid flow in the shell side due to the perpendicular arrangement of the outlet. The outlet channel shows no distinct influence on the lumen to shell sided permeation. Only the flow is directed to the outlet in section C-C.

3.6. MRI velocimetry - dead-end pure water forward permeation

Next, the CFD simulations are compared and validated by flow-MRI measurements. Figure 8 shows the axial and radial velocity magnitude in the multibore membrane during dead-end pure water inside-out filtration in which arrows indicate the direction of flow. Potting of the membrane inside the module and its swelling behavior cause a bent of the membrane towards one side (here towards the left side). The results are comparable to the CFD simulation of the sagging membrane in Figure 4.

The axial velocity magnitude at the shell side is highest at the maximum distance between the membrane and inner tube wall, caused by small local pressure differences on the lumen side [33]. This is in good agreement with the CFD results shown in Figure 3 and Figure 4.

Fluid inside the bore channels forms parabolic laminar profiles similar as it can be seen in the CFD results. Contrary to the CFD simulations, the flow is distributed unevenly within the seven bores. Thus, the bores contribute unequally to the filtration process. The velocity magnitude of the upper bores is 8 % greater compared to the bottom ones. One possible reason might be based on small differences in the shape of each channel.

The illustration of the radial velocity magnitude in Figure 8 contains additional arrows to depict the direction of the radial velocities. A secondary flow field forms around the membrane from the shortest distance between membrane and inner tube wall to the largest distance. This behavior can also be observed in the CFD simulations of the excentric membrane (Figure 3) and the sagged membrane (Figure 4). Secondary flow fields arise when the membrane is not situated concentrically within the housing due to local pressure differences.

In flow-MRI measurements, radial velocities in all bore channels are oriented in one direction. In comparison, this effect is only observed for the CFD study of the sagged membrane (A-A, Figure 4) and not for the study of the excentrically positioned membrane. The sagging between two fixed positions occurs due to the manual manufacturing of the module and the swelling behavior of the polymer membrane.

The shown images of both, measured and simulated flow field, are averaged over a thickness of 1 cm. The net velocity follows the direction of the sagging membrane that is not

parallel to the measurement axis of the NMR tomograph. Therefore, averaging results in flux in one direction in every bore. However, this phenomenon also occurs in an infinitesimal slice in the CFD simulation.

Due to higher pressures inside each bore channel, flux that has left one bore does not re-enter another bore. Contrary to the CFD simulations, water flows through the porous matrix as opposed to the outer bore channels, being the path of least resistance. This can be explained by the skin formation around the bores and the gradual porosity of the polymeric matrix. The results of the CFD simulation, however, show the behavior of ceramic multibore membranes with only little gradients in porosity.

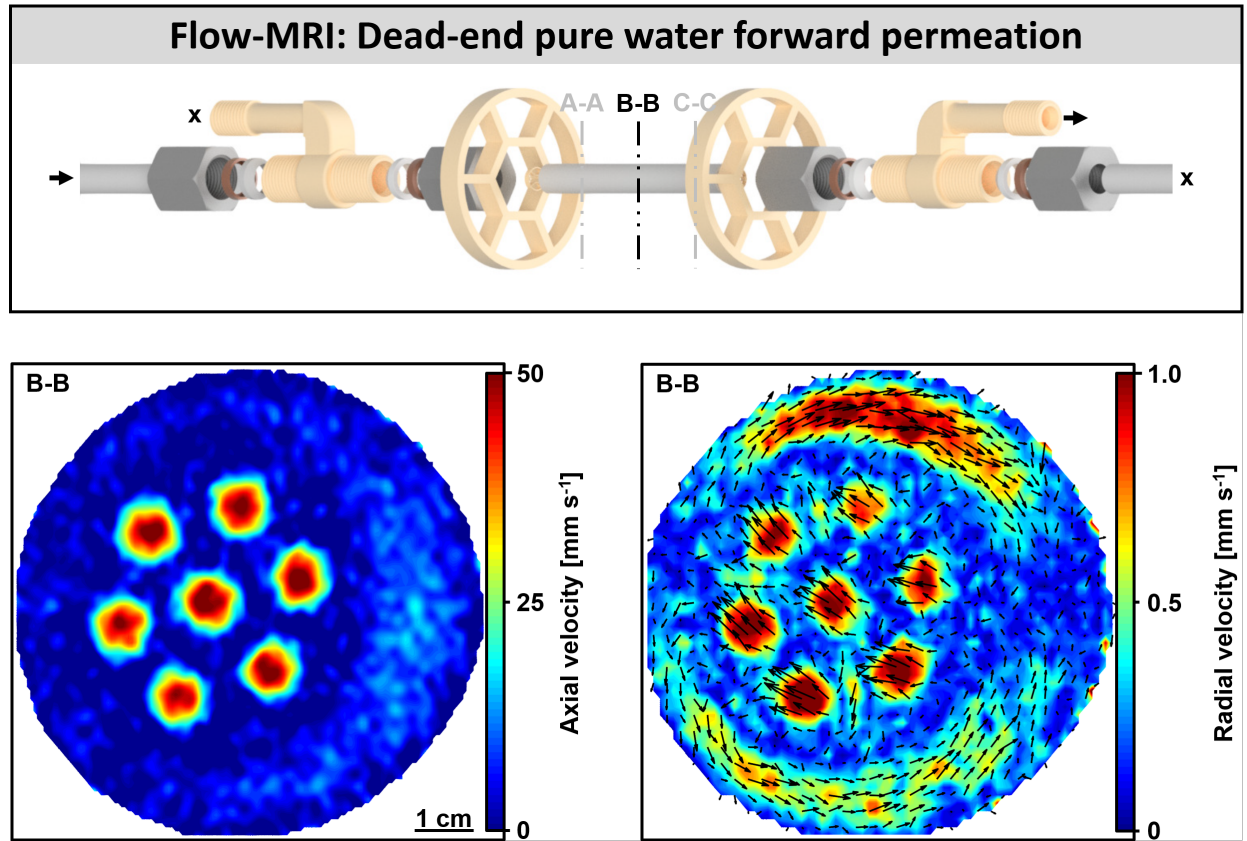


Figure 8: MRI velocimetry - pure water dead-end filtration: Shown is the slice in the center of the module. The characteristic axial (left) and radial (right) flow pattern match the CFD simulations of the sagging membrane in the module. The sagging of the membrane introduces secondary flow fields in the lumen channel and a directed annular gap flow on the shell side.

3.7. MRI velocimetry - dead-end pure water backwash

Additionally to the pure water filtration measurements in dead-end mode operation, we investigated reversed flow (backwash) filtration, which is typically applied to dislodge any filtration deposits from the membrane surface to recover membrane permeability. Figure 9 depicts axial and radial velocity magnitudes in the multibore membrane during dead-end backwash filtration at positions A-A and B-B. Images of the radial velocities contain a sketch of the multibore membrane in white color. Backwash flow enters through the T-connection next to the reject outlet and leaves the module at the back side as can be seen at the top of Figure 9.

During backwash, the axial velocities on the shell side behave similarly to the velocities observed in the dead-end filtration mode in section 3.6. Here, negative velocity values indicate a flow outwards of the measured plane while positive values represent a flow into the plane. Most of the fluid flows through the part with the greater space between membrane and tube wall. At positions further away from the backwash water inlet (from B-B towards A-A), velocity magnitude decreases in both radial and axial direction. This is an indication for lower backwash effectiveness in real applications using this module geometry. Axial velocities in the lumen decrease from B-B to A-A by up to 25 % and are unevenly distributed. This may also cause an uneven cleaning performance per bore in a multibore membrane. Using flow-MRI, we can show that local fluxes vary over the membrane length influencing the backwash flux locally [22].

In contrast to the flow-MRI measurement shown in Figure 8 and all other CFD simulation results, shell side radial velocities in Figure 9 at position A-A flow in a different direction. Here, the secondary flow field is directed towards the area with the smallest distance between membrane and outer tube wall. However, at position B-B radial shell velocities change their direction towards two o'clock. A reason could be the change in position of the membrane in this plane. Additionally, the overall velocities are higher suggesting a potentially higher cleaning efficiency.

Backwash flow preferentially enters the membrane from the bottom shell side. It splits and flows into the bottom bores and partly through the porous membrane structure between

the bottom bores to reach the middle and the top bores. Besides, we observe flux pathways from outer bores towards the center bore as suggested by Zhu et al. [20].

All in all, these measured backwash flow velocities allow us to indicate an uneven distribution of fluid flow over the different bores. Using this module design, radial and axial velocities decrease moving away from the backwash inlet, which results in a lower force for dislodging particles and carry them into the bulk phase. Performing a backwash alternating from two sides, front and back part of the module, or moving the backwash inlet into the middle of the module could be beneficial to the permeability recovery of such membrane modules. The skin of the bores is the part of the membrane with the highest local flow resistance. The porous structure and outer skin represent a comparable lower resistance to the backwash flux. Contrary to what one would expect, Figure 9 shows that the center bore is not only reached by the backwash water but shows the highest velocities measured. The longer distance between shell and center bore compared to the distance between shell and outer bores does not negatively impact the backwash effectiveness in the center bore.

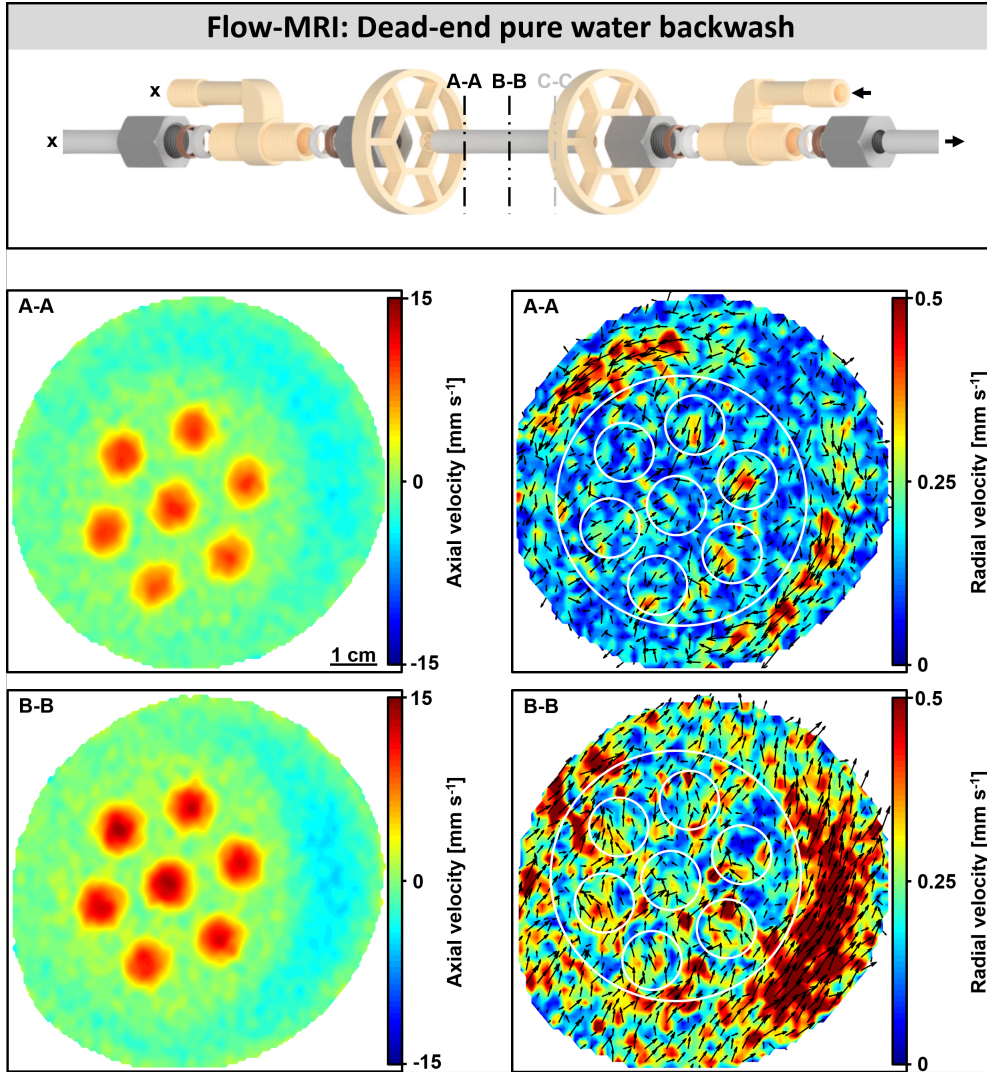


Figure 9: MRI velocimetry - pure water dead-end backwashing: Shown are the backwashing streams at two different positions within the membrane in axial (left) and radial (right) direction. The axial velocity indicates an uneven distribution of the flow between the lumen channels. The central lumen channel is reached during backwashing. The shell sided radial velocities suggest infiltration of the bottom bores first.

3.8. MRI velocimetry - dead-end silica filtration and backwash

Next, the fouling behavior is investigated by MRIs of the multibore membrane module. An aqueous slurry of silica particles is filtered for 60 min as described in Section 2.4. Two distinct cross-sections (A-A and B-B) are displayed in Figure 10 after the filtration with silica particles and after one and two backwash cycles, respectively.

It is possible to distinguish between shell side, porous membrane and silica particles inside the bores. The distribution of deposited particles looks similar at both cross-sectional planes after filtration of the silica particle suspension. The three bores at the bottom are entirely blocked at position A-A and partly blocked at position B-B. A thicker dark ring appears in the middle bore which indicates deposited particles at the membrane wall. After fouling with silica particles, we applied a first constant backwash flux of 180 LMH enabling the removal of particles in bottom bores. The silica particles deposited in the middle bore remain. Images after a second backwash step at 360 LMH show no significant difference to the images after the first backwash step. From this analysis, one might conclude that a backwash flux higher than 180 LMH is not more effective in removing the silica particles.

To evaluate the state of the bore channel during a whole filtration and backwash cycle, we performed dead-end pure water flow measurements after each step (fouling, first backwash, second backwash) at position B-B. Figure 11 shows the velocity magnitudes in axial direction before and after silica filtration and after each backwash step. Although the MRIs indicate a recovered membrane after the first backwash step at 180 LMH, the measurements of the flow-MRI velocimetry fields indicate otherwise. Before silica filtration, comparable flow profiles in each bore build up and are evenly distributed (small differences as explained in section 3.6). Due to silica particle deposition (Figure 11 - after silica filtration) flow in three bores vanishes completely. The MRI at position B-B after filtration of silica particles implies a free bore at the top right location. Still, nearly no flow is observable in this bore. This indicates a blockage in front of position B-B, between feed inlet and measured plane.

Free bores have to compensate flow for the blocked ones because of the dead-end filtration mode. Thus, flow is not distributed equally within all empty bores. The velocity in the top bore rises by 335 % to keep up the same filtration performance as before fouling.

Backwash with a flux of 180 LMH successfully removes deposited particles at this position (see Figure 10). However, velocity flow fields give a contrary picture. Except for the bottom bore that starts working again after the first backwash step, the other three bores are still inactive. To remove all particles between feed inlet and position B-B, we applied a second backwash step at 360 LMH. Now velocity profiles in all bores are visible. However, one bore still seems to be more blocked than the others. The higher velocities in the top bore indicate a not fully recovered membrane.

The combination of flow-MRI and MRIs visualize the dynamic interplay between particle deposition, backwash, and flux transport pathways that all together is difficult to predict. Using flow-MRI, we can determine the required backwash flux for a specific membrane module length to recover its filtration performance. Additionally, to improve the backwash performance it could be favorable to backwash alternating from two sides, or from the middle of the module or to have flow distribution channels (like a perforated permeate tube in commercial modules with a plurality of multibore membranes) to assist backwash effectiveness.

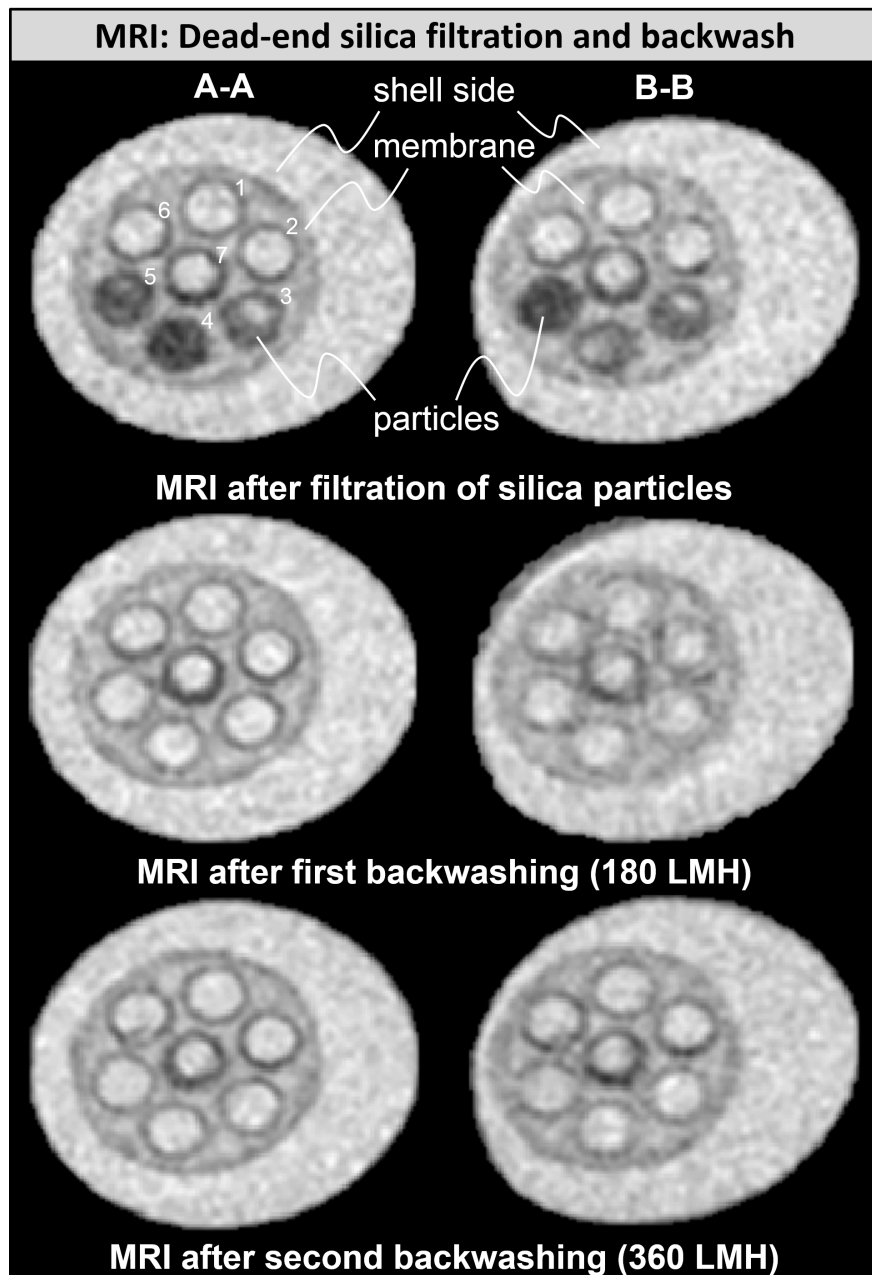


Figure 10: MRI - silica filtration: Images show the deposition of silica particles after filtration and after two subsequent backwash steps. The images of the backwash show that all bores are presumably free of deposits.

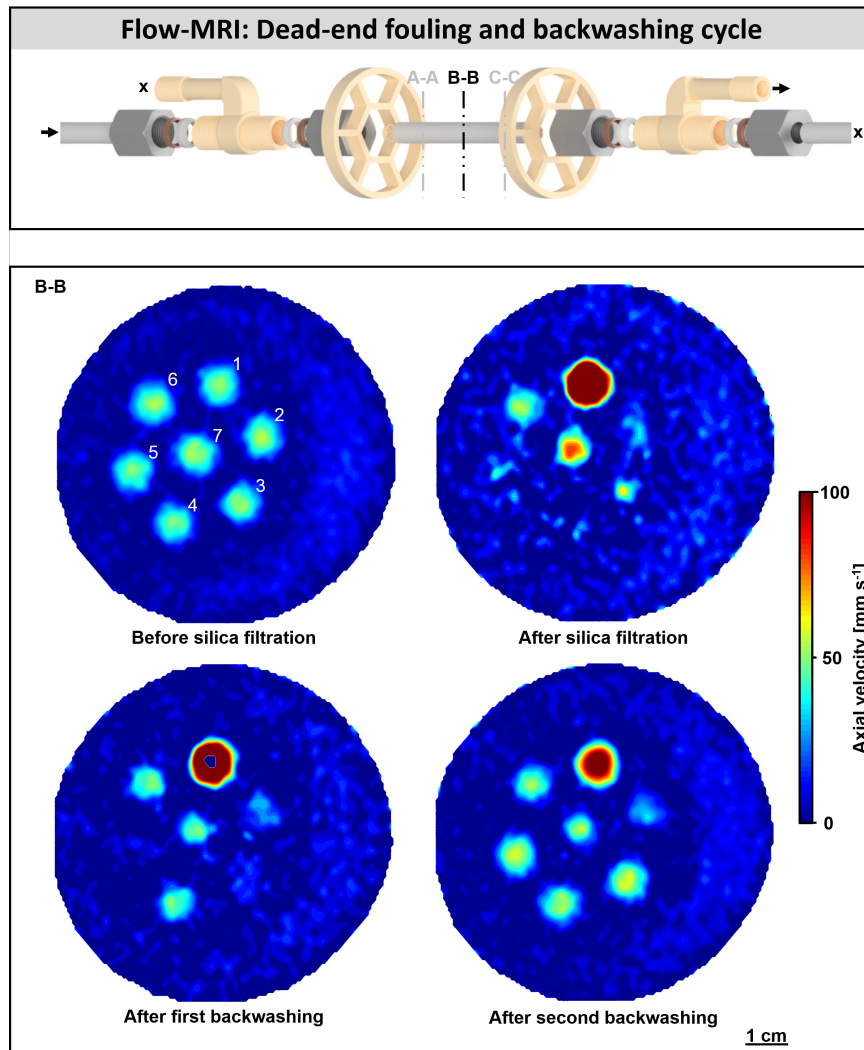


Figure 11: MRI velocimetry - silica dead-end filtration: Before filtration, all lumen channels are available for filtration. The foulant deposits in different positions. As opposed to the MRI images, not all lumen channels are sufficiently cleaned after the first backwash step. The channels must be recovered with a higher backwash flux.

3.9. MRI velocimetry - cross-flow silica filtration

In this study, in contrast to all other measurements and simulations before, Figure 12 shows results of silica fouling experiments in a cross-flow set-up with a constant flux of 100 LMH. In this time-dependent fouling experiment, images were taken every 15 min consecutively at position B-B.

Silica particles start to deposit in the top right corner after 15 min of filtration in all seven bores. The cake layer grows circularly and towards the middle of each bore for about 60 min. Since there is no significant increase in cake layer thickness from 60 to 120 min, the maximum thickness is presumably reached after 60 min of filtration. However, from 60 min on, a denser ring develops at each bore wall. A deposition covering the whole circumference becomes more visible. As the deposition is higher in the top right, the preferential flow finds its way at lower resistant areas away from the top right, and the deposition density grows in a ring away from the top right. The bores remain open due to the cross-flow mode of operation.

A possible explanation for this behavior is the position of the module's outlet position. It is situated in the top right corner. Due to the constant flux of 100 LMH, drag forces may force the silica particles to deposit at this position shown in Figure 12. Here, drag forces are higher compared to gravitational forces.

In the future, a faster imaging sequence (e.g., FLIESSEN [39]) could clarify where the deposition starts precisely. The SPIN ECHO sequence used in our study averages the magnetic resonance signal over 15 min of filtration and is too slow to resolve the initial deposition phenomena.

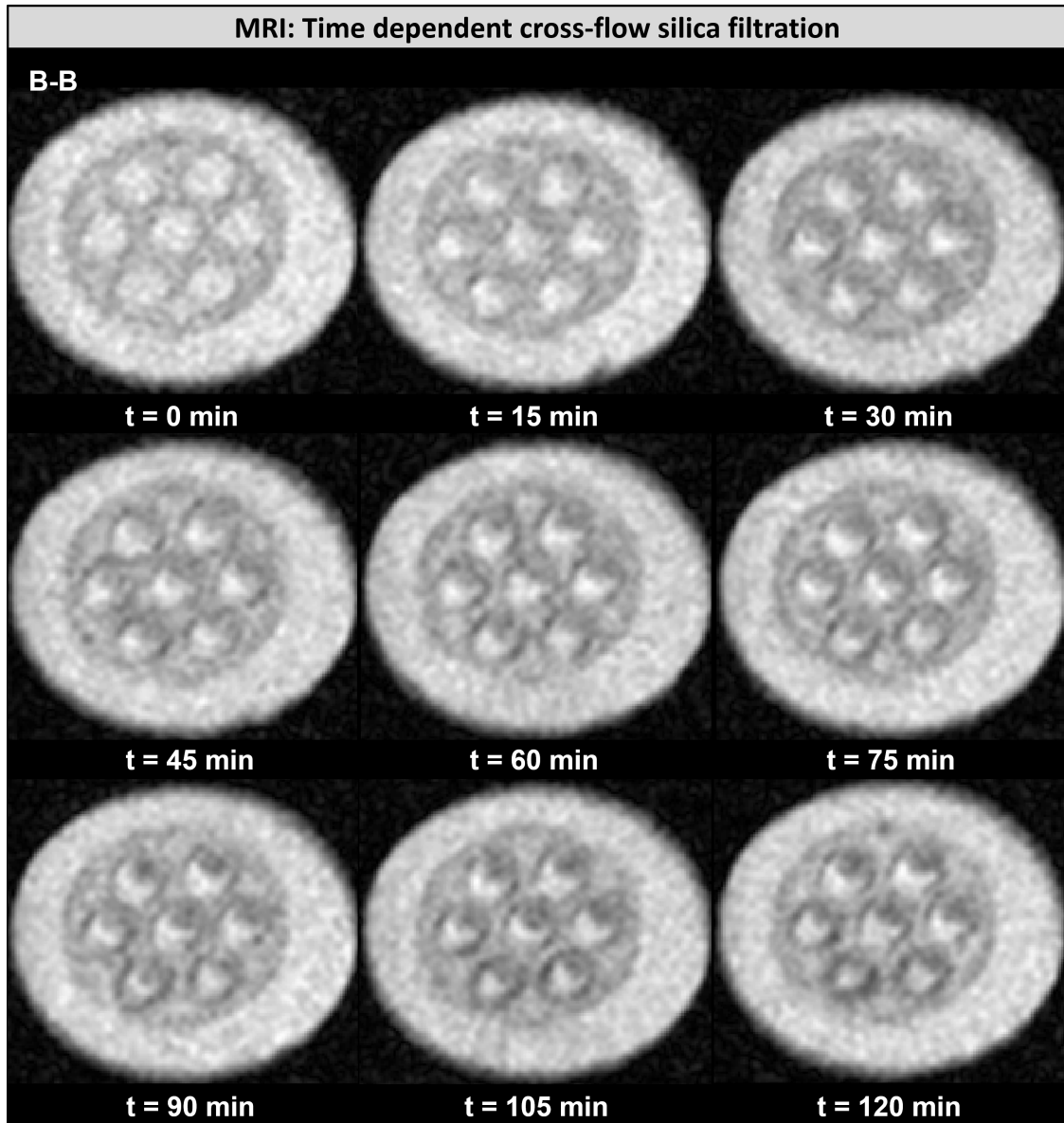


Figure 12: MRI - silica cross-flow filtration: Shown are images of different time steps during cross-flow filtration. The cake layer build-up is different compared to dead-end filtration. Drag forces are presumed to be the reason for the cake layer deposition in the upper right corner of each bore.

4. Conclusion

In this study, we investigate the influence of the hydrodynamics on shell and lumen side flow and pressure communication during membrane operation. MRI, flow-MRI, CFD simulations, and permeation experiments inside a custom made multibore single membrane module evaluate the pathways of filtration and backwash flows. This combination helps to deeper understand (a) flow phenomena inside multibore membranes in dead-end filtration and backwash mode (b) the membrane’s interaction with its surrounding inside the module because of a non-ideal membrane positioning and (c) particle deposition behavior during dead-end and cross-flow filtration mode.

First, CFD simulations are used to mimic challenges within the production process of building a membrane module. In a perfectly aligned concentric membrane module in dead-end inside-out filtration mode, the volume flow decreases in the lumen channels and increases in the shell side according to the membrane’s permeation properties. The fluid flow permeates with a directed radial flow path through the membrane. The resulting flow field shows no secondary flow field on the shell side. This changes when the positioning of the membrane is changed.

- An excentric position of the membrane towards the inner tube wall of the module creates a secondary flow field on the shell side due to different distances between membrane and tube wall and thus, small local pressure differences.
- A sagging membrane due to swelling during permeation and a fixed end position result in a radial flow field formation on shell and lumen side. On the shell side, similar phenomena as in an excentric position arise. Radial secondary flow in bore channels in the bent direction is observed.
- A permeate outlet perpendicular to the multibore membrane orientation only influences the shell side flow in the vicinity of the outlet. The directed flow through the membrane is not affected.

- The assumption of the inlet flow field entering the membrane equally distributed in each bore channel is not necessarily valid. These differences influence the availability of bore channels for filtration. Future investigations should focus on the effect of heterogeneous membrane properties (modeling the local porosity and local permeability of the porous membrane matrix) as well as inlet flow designs of membrane modules.
- A damaged separation skin does not have a significant negative influence on the flow field and performance of the membrane. The flow diverts insignificantly from its original path. The membrane structure protects the inner separation skin. Hence, a decrease in selectivity is unlikely.

Flow-MRI measurements positively validate the CFD simulations. The infinitesimal CFD simulation slices must be averaged to a slice of 1 cm in correspondence to the MRI slices to successfully match and compare the results.

Second, heterogeneous fouling of the membrane with silica particles occurs due to the geometry induced flow patterns. The foulant deposits more strongly on distinct positions. This leads to sequential fouling of bore channels. Opposed to the dead-end flow arrangement, we showed that the outlet position of a membrane module has an influence on the particle deposition during cross-flow filtration. Drag forces cause particles to deposit on the top right corner. MRI and flow-MRI measurements in the subsequent backwash experiments reveal that (a) the backwash flux does reach the center bore despite a longer pathway and (b) effective backwash relies on a backwash flux large enough to reach every bore channel. Even two backwash steps at 180 LMH and 360 LMH were not sufficient to recover the membrane completely. Even if it appears that the membrane is recovered completely, there may still be deposits on the membrane surface (irreversible fouling). Dead-end filtration fouling reveals a significantly different fouling behavior as compared to cross-flow. This should be considered when choosing an operation mode for the filtration of high solid content solutions.

This study reveals the importance of the module geometry on the flow field during membrane permeation. Unavoidable non-idealities of membrane positioning during construction of the module influences the filtration performance, fouling behavior and backwash capabil-

ities. Combining MRI, flow-MRI, CFD simulations, and permeation experiments pioneers the pathway to find strategies to optimize membrane module design and operation, including strategies on how to best recover the membrane performance.

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