MATHEMATICAL MODELING OF CHANNEL – POROUS LAYER INTERFACES IN PEM FUEL CELLS

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ABSTRACT

In *proton exchange membrane (PEM) fuel cells*, the transport of the fuel to the active zones, and the removal of the reaction products are realized using a combination of channels and porous diffusion layers. In order to improve existing mathematical and numerical models of PEM fuel cells, a deeper understanding of the coupling of the flow processes in the channels and diffusion layers is necessary.
After discussing different mathematical models for PEM fuel cells, the work will focus on the description of the coupling of the free flow in the channel region with the filtration velocity in the porous diffusion layer as well as interface conditions between them.
The difficulty in finding effective coupling conditions at the interface between the channel flow and the membrane lies in the fact that often the orders of the corresponding differential operators are different, e.g., when using stationary (Navier–)Stokes and Darcy's equation. Alternatively, using the Brinkman model for the porous media this difficulty does not occur. We will review different interface conditions, including the well–known Beavers–Joseph–Saffman boundary condition and its recent improvement by Le Bars and Worster.

1. INTRODUCTION

Numerical simulation of coupled flows in plain and porous media is essential for many industrial and environmental problems: *proton exchange membrane (PEM) fuel cells*, flow through (oil) filters [17], contaminant transport from lakes by groundwater, $\mathrm{CO_2}$ sequestration in the subsurface, salt water intrusion, etc..
In this work we will focus on coupling conditions between the pure liquid flow and the flow in the porous media. Coupling conditions are well studied only in the simple case of parallel flow over a porous media.
In general, we distinguish two types of PEM fuel cells: H_2 PEM fuel cells ($\mathrm{H_2}$PEMFC) driven by gaseous hydrogen, and *direct methanol fuel cells (DMFC)* operating on methanol in an aqueous solution. Both anode and cathode consist of supply channels, a porous diffusion layer and an active zone. They are connected by a proton conducting membrane. For details we refer the interested reader to [11], [12].
The most important chemical reactions in PEM fuel cells are

	DMFC	$\mathrm{H_2}$PEMFC
Anode	$CH_3OH + H_2O \to CO_2 + 6H^+ + 6e^-$	$3H_2 \to 6H^+ + 6e^-$
Cathode	$\frac{3}{2}O_2 + 6H^+ + 6e^- \to 3H_2O$	

Consequently, in an $\mathrm{H_2}$PEMFC, ideally, the anode contains only hydrogen, while the cathode contains a mixture of liquid water, water vapour and oxygen resp. air. While for an optimal supply of oxygen, it is desirable to keep the amount of liquid water at the cathode minimal, the optimal proton conductivity of the membrane is reached only if it contains enough water. Hence, the water management is an essential issue.
In a DMFC, which is operated on an aqueous solution of methanol, we always can assume that the membrane is wet enough to ensure high conductivity. However for this type of fuel cell, methanol permeation through the membrane, leading to a parasitic reaction on the cathode side, is a key problem. Another problem is clogging of the anodic channels by $\mathrm{CO_2}$ bubbles.
In spite of our remark on the validation of current coupling models, most models either focus on the processes in the membrane electrode assembly (MEA), or in the fluidic channels, simplifying the other process, respectively. A further complication comes from the fact that in both cases, the general process includes two phase flow of a fluid and a gas mixture.
To start with, this paper discusses various options for coupling algorithms between porous transport layers and fluid channels in the case of one-phase flow under the aspect of usefulness in the context of PEM fuel cells.

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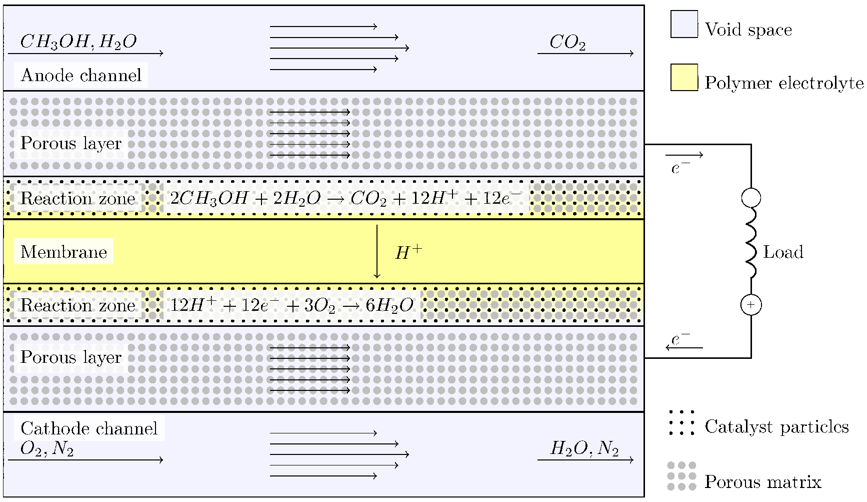

Figure 1: Schematic of a Direct Methanol fuel cell with anodic and cathodic interfaces between porous transport layers and supply channels.

2. NUMERICAL ALGORITHMS

All numerical algorithms for solving the coupled system of free fluid and porous media can be traditionally classified into two groups of methods.

The first group of methods uses *different equations in different subdomains*, e.g., the Navier–Stokes equation in the liquid region and the Darcy / Brinkman model in the porous zones and couples them through suitable interface conditions. These kind of algorithms are (naturally) based on *domain decomposition* techniques [9]. The advantage of this approach is that one can use existing algorithms and software for solving Navier–Stokes equations and porous media flows. However, the problem of this two–domain approach lies in coupling the conservation equations in both regions through the use of appropriate boundary conditions at the interface.

The second group consists of those algorithms, that solely uses *one system of equations in the whole domain* (Navier–Stokes–Brinkman system) obtaining the transition between both fluid and porous regions through continuous spatial variations of properties ('single-domain approach'). Usually, like in most commercial CFD software (e.g. Star-CD, etc.), the Navier–Stokes–Brinkman system is solved by algorithms developed for the Navier–Stokes system modified such that the main term describing the flow through the porous media is treated explicitly. Again, in some cases the algorithms converge very slowly (or even fail to converge).

3. MATHEMATICAL MODELS

In this section we will review some adequate (macroscopic) stationary mathematical models for the flow in each subdomain. In the following Ω_f denotes the pure fluid domain and Ω_p is the porous region (membrane). It is essential to recognize that the velocity and pressure variables in Ω_f and Ω_p have different meanings but we will use the same notation for both. While in the fluid part $\mathbf{u}$ and p denote the usual velocity and pressure, in the porous media $\mathbf{u}$ and p are spatially averaged (over a representative elementary volume (REV)) microscopic quantities. The velocity in the porous domain Ω_p is often referred to volumetric flux density, *Darcy velocity* or *filtration velocity*.

3.1 Models in the Free Fluid Region

The free flow in the fluid region Ω_f is modelled by the laminar *incompressible isothermal Navier–Stokes equations* (or by Stokes equations, i.e. neglecting the convective term $(\rho\mathbf{u}\cdot\nabla)\mathbf{u}$ in the case of creeping flows):

$$-\mu\Delta\mathbf{u} + (\rho\mathbf{u}\cdot\nabla)\mathbf{u} = \mathbf{f}_{\text{NS}} - \nabla p, \qquad \nabla\cdot\mathbf{u} = 0 \qquad \text{in } \Omega_f, \tag{1}$$

where

$$\Delta\mathbf{u} = [\Delta u^1, \ldots, \Delta u^N]^\top, \qquad (\mathbf{u}\cdot\nabla)\mathbf{u} = \left[\mathbf{u}\cdot\nabla u^1, \ldots, \mathbf{u}\cdot\nabla u^N\right]^\top \text{ with velocity } \mathbf{u} = (u^1, \ldots, u^N)^\top,$$

for dimensions $N = 2, 3$. In eq. (1) p is the pressure, μ the fluid viscosity and ρ denotes the density.

3.2 (Macroscopic) Models in the Porous Media

Usually the saturated flow in the porous media Ω_p is described by the famous *Darcy model* discovered 1856

$$\mu\mathbf{K}^{-1}\mathbf{u} = \mathbf{f}_\mathrm{D} - \nabla p, \qquad \nabla\cdot\mathbf{u} = 0 \qquad \text{in } \Omega_p, \tag{2}$$

with μ the fluid dynamic viscosity, $\mathbf{K}$ permeability tensor of the porous medium and $\mathbf{f}_\mathrm{D}$ is a force term (e.g. the gravity). In eq. (2) $\mathbf{u}$ denotes the volumetric average of the velocity and p is the average of the pressure.

An extension of this model (2), the *Brinkman model* [4], is usually used in order to account for the *high porosity* of the porous media or to impose no–slip conditions on solid walls:

$$-\nabla\cdot(\mu_\mathrm{eff}\nabla\mathbf{u}) + \mu\mathbf{K}^{-1}\mathbf{u} = \mathbf{f}_\mathrm{B} - \nabla p, \qquad \nabla\cdot\mathbf{u} = 0 \qquad \text{in } \Omega_p, \tag{3}$$

where $\mu_\mathrm{eff} = \mu/\phi$ is the *effective viscosity* of the fluid in Ω_p and ϕ denotes the porosity of the porous media. In order to decide which model is adequate there exists a *rule of thumb*: the Brinkman model is used if the Reynolds numbers $\mathrm{Re} = \rho U L/\mu$ of the corresponding free flow is greater than 10. Here U and L are characteristic values for the velocity and the length of the whole problem.

4. INTERFACE CONDITIONS BETWEEN FLUID AND POROUS MEDIA (DARCY)

In this section we will discuss the aspects of proper interface conditions between different media. We consider in the sequel the (Navier–)Stokes equations (1) in the free fluid region Ω_f, coupled across an interface with the Darcy equation (2) in the porous medium Ω_p. This is the most common and mathematically the most difficult case, since these two models are completely different systems of PDEs. Hence, it is not clear what kind of conditions should be imposed at the interface Σ between Ω_f and Ω_p. The *classical coupling conditions* for an inviscid fluid are the continuity of the pressure and the continuity of the normal velocities at the interface. For a viscous flow, one would assume additionally the vanishing of the tangential velocity at the interface Σ.

Now, if the interface would be a boundary, then in the fluid part the system needs, e.g., a prescribed velocity (N conditions) and the equation in Ω_p must be supplied with a given pressure or normal flux (1 condition). For coupling Darcy's model (2) and Stokes equation (1) some (well–known) interface conditions are needed to obtain a well–posed problem. Usually, these interface conditions describe the *continuity of the mass flux*

$$\mathbf{u}\cdot\mathbf{n}|_{\Sigma_p} = \mathbf{u}\cdot\mathbf{n}|_{\Sigma_f}, \tag{4}$$

where Σ_p, Σ_f is the same interface Σ seen from porous and fluid parts. Let us note that eq. (4) is not sufficient to calculate the flow in Ω_p, since the flux is yet unknown.

4.1 The Interface Conditions of Ene, Levy and Sanchez–Palencia

For the choice of further interface conditions we need a *classification of the flow*. This was done 1975 by Ene, Levy and Sanchez-Palencia [10], [27]: they distinguished two principally different cases of flow situations named in [22]:

npf (near parallel flow): the velocity in Ω_f is significantly larger than the filtration velocity in Ω_p. The pressure gradients are of similar magnitude in both subdomains.

nnf (near normal flow): the velocities are of similar magnitude in both subdomains and the pressure gradient in Ω_f is significantly smaller than in Ω_p and nearly orthogonal to Σ_p.

Depending on this classification different interface conditions additional to (4) were proposed in [10], [27].

4.1.1 The Case of 'Near Parallel Flow'

For the case of *near parallel flow* Ene, Levy and Sanchez-Palencia [10], [27] suggested to use the conditions

$$\text{npf:} \qquad \mathbf{u}|_{\Sigma_f} = 0, \qquad p|_{\Sigma_f} = p|_{\Sigma_p}. \tag{5}$$

The first condition in (5) originates from the *continuity of velocity* across the interface where the filtration velocity in Ω_p is neglected. Note that this simplification allows to compute (in principle) the flow solely in the domain Ω_f. Hence, the pressure field is known in the fluid part Ω_f and via the *continuity of pressure* condition in (5) also on the interface Σ_p. Afterwards, the pressure field in the whole porous media can be determined by solving the elliptic Darcy equation (2) with prescribed pressure values on Σ_p. Doing so, one obtains a nonzero normal component of the filtration velocity in Ω_p, i.e., the mass flux condition (4) holds only roughly.

4.1.2 The Case of 'Near Normal Flow'

For this case of *near normal flow* the authors proposed in [27] the interface conditions

$$\text{nnf:} \qquad p|_{\Sigma_p} = C, \qquad \mathbf{u} \cdot \tau_j|_{\Sigma_f} = 0, \qquad j = 1, \ldots, N-1. \tag{6}$$

C denotes an a-priorily unknown constant and τ_j are the orthogonal unit tangent vectors to the interface Σ_f. Since the pressure is usually defined only up to a constant, it is often convenient to assume that the pressure p at the porous interface Σ_p takes a certain (arbitrary) constant value C. Doing so, one neglects the dependence of $p|_{\Sigma_p}$ on the fluid flow in Ω_f (compared to the strong dependence in the porous media Ω_p). For a chosen constant C first the flow in Ω_p can be determined and then the problem in the fluid domain is solved using the mass flux condition (4) and the second condition in (6) for the tangential velocity components.

4.2 The Beavers–Joseph Interface Condition

In 1967 Beavers and Joseph [3] performed several experiments in a fluid channel over a porous media and found out that the mass flux through Ω_f is larger than predicted by the Poiseuille flow (i.e. with no-slip boundary conditions). This flow situation can be classified as a case of *near parallel flow* (cf. Section 4.1.1) with interface conditions (5). Beavers and Joseph explained this observation with a *slip velocity* at the interface and proposed an empirical *slip-flow condition* that agreed well with their experiments (cf. Fig. 2):

$$\frac{\partial u}{\partial y}(x, 0+) = \frac{\alpha}{\sqrt{K}}\big(u(x, 0+) - u_D\big), \tag{7}$$

where u_D denotes the uniform tangential (horizontal) Darcy velocity in Ω_p $(-H < y < 0)$ obtained from the Darcy equation (2) and $u(x, 0+)$ is the tangential velocity component in the fluid region Ω_f $(0 < y < G)$ and K denotes the permeability. This interface condition (7) relates the gradient of the free flow velocity at the interface $y = 0$ to the filtration velocity u_D. The Beavers–Joseph constant α (measured slip coefficient) in eq. (7) only depends on porous media properties. It denotes a dimensionless quantity depending on the material parameters which characterize the structure of the permeable material within the boundary region and its values ranges between 0.01 and 5 [30]. Let us point out that eq. (7) allows for a discontinuity in the tangential velocity, i.e., rapid changes in the velocity in a small boundary layer are substituted through a jump. Using the Beavers–Joseph condition (7) the agreement between measurements from their experiments and the predicted values was quite good, with over 90% of the experimental values having errors of less than 2% [3]. The work of Beavers and Joseph was continued by investigations of Taylor [40] and Richardson [34] and an extension, the *Jones condition*, was proposed in [21].

Later on Payne and Straughan [33] showed the continuous dependence of the solution on the Beavers–Joseph constant α in eq. (7). Moreover, the interface condition eq. (7) was mathematically justified by Jäger and Mikelić [18]. Although this condition is not justified in the general case, it is widely used in practical computations for coupled fluid flows, e.g., in [8], [25], [28], [35], [41] and especially many tests in [6].

4.3 Saffman's Modification of the Beavers–Joseph Interface Condition

In the article [36] Saffman gave a 'theoretical' justification of the Beavers–Joseph interface condition at a physical level of rigor. Moreover, Saffman proposed in 1971 a modification of the Beavers–Joseph law (7): he found out that the tangential velocity on the interface is proportional to the shear stress and proposed a modification of the Beavers–Joseph condition:

$$u(x, 0+) = \frac{\sqrt{K}}{\alpha}\frac{\partial u}{\partial y}(x, 0+) + O(K). \tag{8}$$

While the Beavers–Joseph interface condition (7) couples the fluid velocity in Ω_f with the filtration velocity in Ω_p, the modified eq. (8) (Beavers–Joseph–Saffmann condition) contains only variables in the free fluid domain Ω_f where the filtration velocity is usually much smaller than the slip velocity $u(0)$. If the slip velocity is smaller than the maximal filtration velocity then setting the tangential velocity to zero is a reasonable approximation. We remark that Dagan [7] came in 1979 to the same conclusion. He proposed a so-called Slattery's relation [38] between the pressure gradient and the first two derivatives of the Darcy velocity in order to obtain the condition (8).

5. INTERFACE CONDITIONS BETWEEN FLUID AND POROUS MEDIA (BRINKMAN)

Neale and Nader [31] suggested in 1974 the usage of the Brinkman correction to the Darcy model (3): they proposed to assume continuity of velocity and stress (using μ_{eff}) across the fluid–porous interface since the Stokes and the Brinkman equation are of the same order. Doing so, Neal and Nader obtained in the fluid region the same solution as Beavers and Joseph provided that the slip coefficient is chosen as $\alpha = \sqrt{\mu_{\text{eff}}/\mu}$.
Vafai and Kim [42] constructed 1990 an exact analytic solution for the interface region, including boundary and inertia effects. Later on, Alazmi and Vafai [1] classified and analyzed five primary categories of interface conditions between fluid layer and porous medium (modelled by Brinkman eq. (3)).
In 1992 Sahraoui and Kaviany [37] performed a numerical study and calculated the slip coefficient: they discovered that the Brinkman extension to the Darcy equation does not satisfactorily model the flow field in Ω_p . However, this can be overcome using a variable effective viscosity μ_{eff} in the porous medium.
On the contrary, for the Brinkman model for the flow in Ω_p this ambiguity does not occur. In this case, the equations in the porous media Ω_p and equations in the fluid region Ω_f are of the same type. Two types of coupling conditions can be found in the literature. The more common choice are conditions of *continuous velocity* and *continuity of the normal component of the stress tensor*

$$\mathbf{u}|_{\Sigma_p} = \mathbf{u}|_{\Sigma_f} \tag{9}$$

$$\mathbf{n} \cdot (\mu_{\text{eff}} \nabla \mathbf{u} - p\mathbf{I})|_{\Sigma_p} = \mathbf{n} \cdot (\mu \nabla \mathbf{u} - p\mathbf{I})|_{\Sigma_f}, \tag{10}$$

where Σ_p, Σ_f is the same interface Σ seen from porous and fluid parts. Such conditions would naturally arise, if for some reasons (e.g. in the domain decomposition approach), the fluid region is divided into subdomains, where the Navier–Stokes equations are valid. Usually, the condition (9) is the first one out of N conditions on the interface when considering a Stokes–Brinkman system. This approach is used numerically in [17], [24].

5.1 The Stress Jump Conditions of Ochoa-Tapia and Whitaker

Ochoa-Tapia and Whitaker [32] obtained 1995 at the interface continuity of the velocity and the continuity of the 'modified' normal stress by a volume averaging technique of the momentum equations in the interface region. In their analysis they showed that the matching of Stokes equation with the Brinkman model conserves the continuity of velocity but induces a jump in the shear stress. Hence, they proposed additionally to the condition (9), a *stress jump condition* that takes into account the momentum transfer at the interface

$$\frac{\partial u}{\partial y}(x, 0+) - \frac{1}{\phi_p}\frac{\partial u}{\partial y}(x, 0-) = -\frac{\beta}{\sqrt{K}}\, u. \tag{11}$$

Here, β denotes a dimensionless parameter of order one that is defined as a solution of a closure problem. The authors investigated in [32] the conditions (9), (11) in a 1D channel geometry and compared the results with the classical Beavers–Joseph experiment. These boundary conditions proposed in [32] were used by Kuznetsov [23] to compute solutions in channels partially filled with a porous material.
Many attempts have been made to estimate the adjustable jump coefficient β or to obtain an expression for β, depending on the microstructure of the interface region. In [43] the authors derived a stress jump boundary condition at the interface *free of adjustable coefficients*. The associated local closure problems, modelling the microstructure of the interface, determine a mixed stress tensor which is the reason for the jump.
Furthermore, only few authors have studied the physical nature of these jump coefficients. Jamet and Chandesris [19], [20] analyzed the physical meaning based on an upscaling method of the transport equations in the interfacial region. Doing so, they were able to interpret the jump coefficients as surface tension quantities depending linearly on the position of the interface.
In the dissertation of Laptev [24] a new numerical method in 3D using these interface conditions (9), (11) was proposed. Furthermore, the mathematical model of the coupling of Navier–Stokes and Brinkman equations using the stress tensor jump interface condition (11) was validated for a large class of model problems [24].

5.2 The Transition Zone Approach

When studying a Poiseuille flow over a permeable region, e.g., by Chandesris and Jamet [5], in turned out that the sharp interface with its jump conditions is only the limiting case (i.e. an idealization) of a *transition region*, where the physical properties of the medium have a strong but still continuous variations. Actually, this idea goes back to Nield [29]. He proposed 1983 to use a Brinkman equation in the transition region between the fluid and the porous medium modeled by the Darcy equation. This approach was also validated experimentally by Goharzadeh et al. [13]. They found out experimentally that the thickness of the transition region should be of the same order as the grain size of the porous medium.

In 2003, Goyeau et al. [14] studied the momentum balance at the interface of a two-layer system and introduced a heterogeneous continuously varying transition zone between the 'outer' fluid and porous zones. The authors derived an explicit formula for the stress jump coefficient β involved in the momentum transport. However, this approach assumes the knowledge of the spatial dependence of the effective quantities in the region around the interface.

Chandesris and Jamet [5] solved in 2006 the problem in this transition zone using the technique of *matched asymptotic expansions*: they obtained an explicit representation of the stress jump coefficient in the transition zone depending only on the parameters of the porous medium (permeability and porosity).

Recently, Hill and Straughan [15] considered a *three-layer* constellation: a free fluid interfacing a Brinkman-type porous transition layer, which overlies a porous medium modelled by the Darcy eq. (2). The authors discovered two instability modes that depend on the ratio of the thickness parameters of the different layers.

5.3 The Interface Conditions of Le Bars and Worster

Recently, Le Bars and Worster [26] considered special 'analytically tractable' cases for the one-domain approach with the Brinkman model for the porous medium. The authors compared their findings with the two-domain approach of Section 4 using the Darcy equation and its previously proposed interface conditions, especially the Beavers–Joseph condition (7). Le Bars and Worster considered the Brinkman equation in the configuration studied by Beavers and Joseph, and found a new condition at the fluid-porous interface

$$u(x, -\delta+) = u_D(x, -\delta), \qquad \text{with } \delta = c\sqrt{K}, \tag{12}$$

where c is a constant of order 1. They defined a *viscous transition zone* inside Ω_p, where the Stokes equation still applies up to a depth δ, and imposed continuity of pressure and velocities (9) at the position $y = -\delta$ (cf. Fig. 2). Here, δ denotes the characteristic size of this transition zone (a few pore lengths). Using this new condition (12) the computed values have a (slightly) better coincidence with the experimental values of Beavers and Joseph.

Let us remark that the authors of [14], [26] found good agreement between the single-domain approach of Section 5 and the two-domain approach of Section 4. However, this can be explained by the special configurations, cf. [16], namely a one-dimensional tangential flow in [14] and a seemingly very small vertical velocity gradient on the interface in [26].

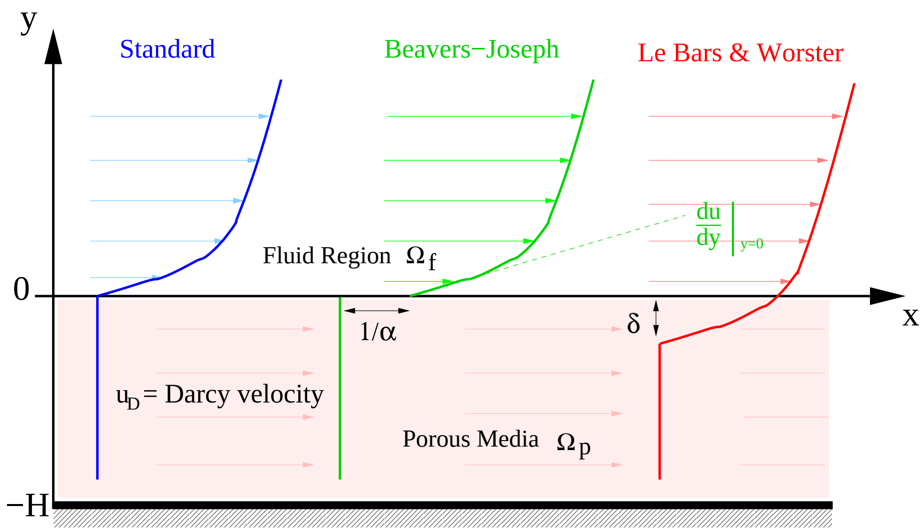

Figure 2: Comparison of Different Interface Models for Porous Media: From left to right: The standard case: no-slip condition on the fluid–porous interface, the Beavers–Joseph condition (7): slip of size $1/\alpha$ on the fluid–porous interface and the Le Bars and Worster condition (12): slip by δ into the porous media.

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