

We want to thank the anonymous referee number 1 for his meaningful and valuable comments which indisputable lead to an improvement of the quality of this manuscript. The following section aims at answering to the referee's comments. Answers are given according to the referee's general comment order.

1. Introduction. The authors almost always point to existing work without giving a brief overview of already existing findings. This should be changed so that the reader understands the knowledge gap that the authors will be filling. The ms is not too long, therefore adding another page or two to the introduction will increase the value of this ms.

We agree to this point and modified the introduction accordingly. We extended the introduction by including the existing findings from the literature. We consider that the section is now more readable and understandable.

The modified introduction is given in the following section. Italic font highlights the parts we have added.

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## **“Introduction**

Understanding the evolution of the fluid-fluid interfaces in two-phase flow in porous media systems is relevant for a series of engineering and technological applications (e.g., CCS, nuclear waste repository, oil recovery, etc.) (Hassanizadeh and Gray, 1990; Joekar-Niasar et al., 2008; Niessner and Hassanizadeh, 2008; Reeves and Celia, 1996). *The quantity of the (typically unknown parameter) interfacial area between two phases restricts kinetic interphase mass and energy transfer (Ahrenholz et al., 2011). In numerical modeling with classical macroscale flow models, these processes are not properly taken into account (Muccino et al, 1998; Ahrenholz et al., 2011). Here, averaged quantities neglecting the interfacial area are often used (Tatomir et al., 2015), which are described to lead to negligence of kinetics of mass transfer and assuming local equilibrium (Niessner and Hassanizadeh, 2009). In order to resolve this, the capillary pressure( $p_c$ )-wetting phase saturation( $S_w$ )-interfacial area( $a_{wn}$ ) relationship using the concept of specific interfacial area per volume described by Hassanizadeh and Gray (1990) can be used.*

Based on an extended form of Darcy's law (considering fluid-fluid friction force and interfacial forces), the mathematical fundamentals of pore scale two-phase flow regarding the  $p_c$ - $S_w$ - $a_{wn}$  relationship, are given in Hassanizadeh and Gray (1980; 1990; 1993), and Niessner and Hassanizadeh (2008). The derivation of this porous media specific  $p_c$ - $S_w$ - $a_{wn}$  relationship can be realized using physical experiments in form of e.g. micromodels as described amongst others in Karadimitiou et al. (2013; 2014), Karadimitriou and Hassanizadeh (2012), and Lenormand and Touboul (1988). Corresponding numerical models are given, among others, in Ahrenholz et al. (2011), and Porter et al. (2009). These specific approaches *succeed in deriving the  $p_c$ - $S_w$ - $a_{wn}$  relationship*, but are based on Lattice-Boltzmann simulations, which are considered as elegant but limited by computational resources (White et al., 2006). Generally, the derivation of this parametric relationship on the pore scale requires a high computational effort as well as a complex numerical model. *Arzanfudi and Al-Khoury (2015) present a FEM- based model that enables to simulate CO<sub>2</sub> leakage through a wellbore. In that one-dimensional approach, the moving CO<sub>2</sub>-water interface is successfully traced using the level set method. Amiri and Hamouda (2013) evaluate two approaches for dynamic interface tracking, the level set method and the phase field method, for two-dimensional modeling of two-phase flow with viscosity contrast in dual-permeability porous medium. Both approaches enable to quantify the specific interfacial area between the fluids. They conclude that the phase field method is more successful in complicated porous media, gives more realistic results regarding pressure gradients and fluid profiles and they observed less computation time compared with the level set method.*

Several experimental and numerical approaches were developed to measure the interfacial area between the two fluid phases. Current research in the field of CCS focuses on the design and development of field investigation techniques allowing short response times in on-site plume monitoring and detection. Detection and quantification of the CO<sub>2</sub>-brine fluid-fluid interface could be realized using Kinetic Interface Sensitive (KIS) tracers as described in Schaffer et al. (2013). Several mathematical models for immiscible two-phase flow and KIS tracer transport in porous media were developed in Tatomir et al. (2013; 2014), and Tatomir et al. (2015) whereas the latter also considers compositional effects. *In the latter, a two-phase four component flow and transport model is realized with a kinetic mass transfer of tracers between the two fluids and taking the dissolution of CO<sub>2</sub> and brine into account.* The  $p_c$ - $S_w$ - $a_{wn}$  relationship is seen as an extension to the standard Brooks-Corey model (Brooks and Corey, 1964).

The modeling approaches by Tatomir et al. (2013; 2014) and Tatomir et al. (2015) require the a priori knowledge of the fluid-fluid-solid system specific  $p_c$ - $S_w$ - $a_{wn}$  constitutive relationship (Tatomir et al., 2013).

The present article presents *the corresponding model developed for deriving this specific  $p_c$ - $S_w$ - $a_{wn}$  constitutive relationship*. In this study, a finite element method (FEM) -based pore-scale model *is used in order to determine the dynamic evolution of the  $CO_2$ -water interface in geometries with gradually increasing level of complexity*. Within this case study, we present the workflow that enables the derivation of  $p_c$ - $S_w$ - $a_{wn}$  relationships of sandstone core sample based model domains. We describe a feasible and relatively simple numerical model that enables the dynamic fluid-fluid interface tracking on  $\mu$ -CT obtained geometries. Initial and boundary conditions as well as spatial and temporal discretization are derived using a novel approach taking simplified idealized geometries based on REV-scale pore geometry statistics into account.

*Beside use as input for the model given by Tatomir et al. (2015), the resulting  $p_c$ - $S_w$ - $a_{wn}$  relationships contribute to designing the KIS tracer laboratory experiments described in Tatomir et al. (2015). Furthermore, the derived relationship aims at improving the characterization the Heletz sandstone reservoir (Edlmann et al., 2016; Niemi et al., 2016; Tatomir et al., 2016)."*

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2. There are numerous commas missing, which should be corrected. The authors are using punctuation in equations, which should be avoided for clarity. Some equations (e.g. (7)) are not referenced, which should be added. Check all "whereas", which should mostly be "where".

It is true that we made these mistakes. We carefully rechecked the spelling and punctuation and addressed the issues the referee is addressing to.

3. Eq. (1). I think there is something wrong in Eq. (1): You are adding nabla u and its transposed form, which does not work. The result is added to  $pI$ , which is a matrix.  $F$  is a vector, so the expressions on the RHS are all vectors? Is this consistent with the LHS?

Yes, we mistakenly formulated a wrong form of the momentum balance of the Navier-Stokes equations (NSE). We particularly thank the reviewer for correcting this mistake and changed the whole momentum balance equation to a form of NSE that is more general (Cengel and Cimbala, 2013). This form is given as equation (1).

$$\rho \frac{Du}{Dt} = -\nabla p + \mu \nabla^2 u + F_{st} + \rho g \quad (1)$$

4. Eqs. (3) and (4). Please clarify in the text before, which equation does what.

Yes, we agree. The modified section can be found in the following paragraph.

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“The change of phase component concentration over time can be described as the product of diffusion coefficient and chemical potential known as Cahn-Hilliard equation (Cahn and Hilliard, 1958). In the present model, this equation is used in the form of equation (3) using the terms mobility  $\gamma$ , predefined interface thickness  $\varepsilon$ , mixing energy density  $\lambda$  and a phase field help variable  $\Psi$  comprising the chemical potential. The latter is calculated according to (4).

$$\frac{\partial P}{\partial t} + u \cdot \nabla P = \nabla \cdot \frac{\gamma \lambda}{\varepsilon^2} \cdot \nabla \Psi \quad (3)$$

$$\Psi = -\nabla \varepsilon^2 \nabla P + (P^2 - 1)P \quad (4)”$$


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5. P15L13.  $u$  is a vector, therefore a “velocity gradient” does not exist. Do you mean velocity difference? How do you calculate that difference when  $u$  is a vector?

Term “velocity gradient” was intended to express the difference in velocities at the wall and the velocity which increases with the distance from the wall. Following Reviewer 1’s observation the term is changed to “velocity difference”.

The comments of anonymous referee number 2 will be addressed soon.