

---

# HyUSPRe

## Hydrogen **U**nderground **S**torage in **P**orous **R**eservoirs

**Validated open-source reservoir modeling software that can simulate coupled flow, geochemical and microbiological processes in a porous reservoir under operational hydrogen storage conditions**

Prepared by: Sebastian Hogeweg, TUC  
Birger Hagemann, TUC  
Leonhard Ganzer, TUC

Please cite this report as: Hogeweg, S., Hagemann, B. & Ganzer, L. (2023) Validated open-source reservoir modeling software that can simulate coupled flow, geochemical and microbiological processes in a porous reservoir under operational hydrogen storage conditions, H2020 HyUSPRe project report. 17 pp.

This report represents HyUSPRe project deliverable number D6.2.

---

## The HyUSPRe consortium



## Funded by



## Acknowledgement

This project has received funding from the Fuel Cells and Hydrogen 2 Joint Undertaking (now Clean Hydrogen Partnership) under grant agreement No 101006632. This Joint Undertaking receives support from the European Union's Horizon 2020 research and innovation programme, Hydrogen Europe and Hydrogen Europe Research.

## Disclaimer

This document reflects the views of the author(s) and does not necessarily reflect the views or policy of the European Commission. Whilst efforts have been made to ensure the accuracy and completeness of this document, the HyUSPRe consortium shall not be liable for any errors or omissions, however caused.

---

## Executive summary

Numerical reservoir modeling is a popular method to simulate (and reproduce) the operational history of an underground gas storage. The underlying aim is to validate and improve these models such that they can be applied to predict the efficiency of the future operation process for the purpose of efficiency optimization and de-risking. This workflow can also be applied to simulate, optimize and de-risk underground hydrogen storage (UHS) operations, the learnings of which are of key importance for the safe, robust, and affordable implementation of UHS on field scale. However, existing commercial simulators developed for hydrocarbon production and CO<sub>2</sub> storage do not properly cover unique processes that may occur when hydrogen is injected into reservoirs, such as bio- and geochemical reactions.

To close this gap, the open-source simulator DuMu<sup>x</sup> was extended with functionality to simulate hydrogen-induced processes and then calibrated to laboratory observations. The newly developed implementation enables users to model two-phase n-component transport of hydrogen-rich gaseous mixtures in porous media coupled with bio- and geochemical reactions. The consumption of hydrogen by methanogenic and sulfate-reducing microorganisms is implemented, which produces methane and hydrogen sulfide (H<sub>2</sub>S) respectively. Furthermore, the pyrite-to-pyrrhotite reduction reaction is implemented, which also generates H<sub>2</sub>S.

Additionally, adaptations within the diffusion modeling based on experimental observations are performed. With the new functionality, this model is able to reproduce laboratory experiments of hydrogen-related processes and can be used to simulate field-scale UHS operational scenarios.

The developed implementation is available as a separate module of DuMu<sup>x</sup> including examples in the Gitlab repository and a persistent dataverse.

---

## About HyUSPRe

### Hydrogen **U**nderground **S**torage in **P**orous **R**eservoirs

The HyUSPRe project researches the feasibility and potential of implementing large-scale underground geological storage for renewable hydrogen in Europe. This includes the identification of suitable porous reservoirs for hydrogen storage, and technical and economic assessments of the feasibility of implementing large-scale storage in these reservoirs to support the European energy transition to net zero emissions by 2050. The project will address specific technical issues and risks regarding storage in porous reservoirs and conduct an economic analysis to facilitate the decision-making process regarding the development of a portfolio of potential field pilots. A techno-economic assessment, accompanied by environmental, social, and regulatory perspectives on implementation will allow for the development of a roadmap for widespread hydrogen storage by 2050, indicating the role of large-scale hydrogen storage in achieving a zero-emissions energy system in the EU by 2050.

This project has two specific objectives. Objective 1 concerns the assessment of the technical feasibility, associated risks, and the potential of large-scale underground hydrogen storage in porous reservoirs for Europe. HyUSPRe will establish the important geochemical, microbiological, flow, and transport processes in porous reservoirs in the presence of hydrogen via a combination of laboratory-scale experiments and integrated modeling; and establish more accurate cost estimates to identify the potential business case for hydrogen storage in porous reservoirs. Suitable storage sites will be identified, and their hydrogen storage potential will be assessed. Objective 2 concerns the development of a roadmap for the deployment of geological hydrogen storage up to 2050. The proximity of storage sites to large renewable energy infrastructure and the amount of renewable energy that can be buffered versus time varying demands will be evaluated. This will form a basis for developing future scenario roadmaps and preparing for demonstrations.

## Document information, revision history, approval status

### Document information

Title:	Validated open-source reservoir modeling software that can simulate coupled flow, geochemical and microbiological processes in a porous reservoir under operational hydrogen storage conditions
Lead beneficiary:	Clausthal University of Technology
Contributing beneficiaries:	-
Due date:	2023.06.30
Dissemination level:	Public
Published where:	www.hyuspre.eu
Recommended citation:	Hogeweg, S., Hagemann, B. & Ganzer, L. (2023) Validated open-source reservoir modeling software that can simulate coupled flow, geochemical and microbiological processes in a porous reservoir under operational hydrogen storage conditions, H2020 HyUSPRe project report. 17 pp.

### Revision history

Version	Name	Delivery date	Summary of changes
V01	S. Hogeweg	2023.06.12	First draft
V02	B. Hagemann	2023.06.22	Review
V03	S. Hogeweg	2023.06.26	Final draft
V04	H. Cremer	2023.06.30	Final edited version

### Approval status

Role	Name	Delivery date
Deliverable responsible:	S. Hogeweg	2023.06.26
Task leader:	S. Hogeweg	
WP leader:	Diego Viesi	
HyUSPRe lead scientist	R. Groenenberg	2023.06.26
HyUSPRe consortium manager:	H. Cremer	2023.06.30

## Table of Content

<b>Executive summary</b> .....	<b>3</b>
<b>About HyUSPRe</b> .....	<b>4</b>
<b>1 Introduction</b> .....	<b>7</b>
<b>2 Implementation into DuMu<sup>x</sup></b> .....	<b>7</b>
2.1 The mathematical model for bio-geo-reactive transport .....	7
2.1.1 Biochemical reactions .....	9
2.1.2 Geochemical reactions.....	10
<b>3 Gitlab repository</b> .....	<b>10</b>
<b>4 Simulation cases</b> .....	<b>11</b>
4.1 Biogeochemical simulation .....	11
4.1.1 Fluid and solid system.....	11
4.1.2 Spatial discretization .....	11
4.1.3 Fluid-matrix interactions .....	12
4.1.4 Initialization .....	12
4.1.5 Operation schedule .....	12
4.2 Cornerpoint.....	12
4.2.1 Fluid and solid system.....	12
4.2.2 Spatial discretization .....	13
4.2.3 Initialization .....	13
4.2.4 Operation schedule .....	13
4.3 Diffusion .....	14
4.3.1 Modeling of binary diffusion coefficients .....	14
4.3.2 Fluidsystem.....	15
4.3.3 Spatial discretization .....	15
4.3.4 Initialization and boundary conditions.....	15
<b>5 Concluding remarks</b> .....	<b>16</b>
<b>References</b> .....	<b>17</b>

## 1 Introduction

Numerical simulations are a popular tool for investigating transport processes in the subsurface to optimize the operation of underground gas storage. Based on empirical data from former field history, the operation of underground energy storage can be optimized. For this purpose, various commercial simulators are available on the market, mainly focusing on hydrocarbons. However, these simulators are often limited in their adaptability and may not cover hydrogen-related processes adequately. For example, biochemical reactions caused by microorganisms such as methanogenic archaea and sulfate-reducing bacteria, and geochemical reactions, can lead to a) changes in the fluid composition that may impact the performance of the reservoir, and b) generation of by-products such as hydrogen sulfide ( $H_2S$ ) that pose a risk for health, safety, and environment can be detrimental to equipment. Modeling these kinds of unique processes can often only be achieved in open-source simulators. In the current study, the open-source simulator DuMu<sup>x</sup> is extended to couple the two-phase n-component transport in porous media with bio- and geochemical reactions. The diffusion model is adapted from the observations in WP4 of the HyUSPRe project to characterize this process accurately. The source code can be obtained from the Gitlab-repository [1] and the persistent dataverse [2].

## 2 Implementation into DuMu<sup>x</sup>

DuMu<sup>x</sup> is in development by the University of Stuttgart (Institute of Modelling Hydraulic and Environmental Systems) since 2007, based on Distributed and Unified Numerics Environment (DUNE), and comes as an additional module to mimic the fluid flow in the subsurface, including chemical reactions. DuMu<sup>x</sup> is the abbreviation for "DUNE for Multi-{Phase, Component, Scale, Physics, ...} flow and transport in porous media" [3], [4]. In general, DuMu<sup>x</sup> does not offer any Graphical User Interface (GUI) and is coded in C++, but a better adjustment of chemical reactions and other effects can be realized. With promising results, [5] implemented the first biochemical reactions caused by microbial activities during UHS.

### 2.1 The mathematical model for bio-geo-reactive transport

The mathematical model for bio-geo-reactive flow is an extension of the mathematical model developed by Hagemann [5], initially developed to simulate microbial processes during UHS. The enhanced model considers two-phase multicomponent reactive transport on the continuum scale (Darcy scale) with an additional compositional solid phase. Here, the molar balance for the fluid components can be expressed as follows:

$$\frac{\partial \phi (\rho_g c_g^k S_g + \rho_w c_w^k S_w)}{\partial t} + \nabla \cdot (\rho_w c_w^k u_w + J_w^k + \rho_g c_g^k u_g + J_g^k) = q^k$$

where  $\phi$  is the porosity,  $\rho$  is the density in  $\text{mol}/\text{m}^3$ ,  $c^k$  is the mole fraction of component  $k$ ,  $S$  is the saturation of the phase,  $u$  is the advective flux in  $\text{m}/\text{s}$ , and  $J$  is the diffusive/dispersive flux in  $\text{mol}/(\text{m}^2 \cdot \text{s})$ . The subscript indicates the phases gas ( $g$ ) and water ( $w$ ), and the superscript  $k$  defines the component.

Furthermore, the mass balance for the solid phase can be expressed as follows:

$$\rho^{k_s} \frac{\partial \phi_s^{k_s}}{\partial t} = q^{k_s}$$

Where  $\rho^{k_s}$  is the molar mass of the solid component  $k_s$  in  $\text{mol/m}^3$ ,  $\phi_s^{k_s}$  is the volume fraction of the solid component  $k_s$ .

On the continuum scale, the advection flow can be described by Darcy's Law for each phase, including the term for gravitational influence:

$$u_i = -\frac{Kk_{ri}}{\mu_i}(\nabla P_i - \hat{\rho}_i g), \quad i = g, w$$

where  $K$  is the absolute permeability in  $\text{m}^2$ ,  $k_{ri}$  is the relative permeability of the phase  $i$ ,  $\mu_i$  is the dynamic viscosity of the phase in  $\text{Pa} \cdot \text{s}$ ,  $\nabla P_i$  is the pressure gradient along the flow path in  $\text{Pa/m}$ ,  $\hat{\rho}_i$  is the phase density in  $\text{kg/m}^3$ , and  $g$  is the gravitational acceleration in  $\text{m/s}^2$ .

The transport due to molecular diffusion is expressed as follows:

$$J_i^k = -\rho_i D_{diff,i}^k \nabla c_i^k, \quad i = g, w$$

where  $D_{diff,i}^k$  is the effective molecular diffusion coefficient in  $\text{m}^2/\text{s}$  and  $\nabla c_i^k$  is the concentration gradient along the flow path in  $1/\text{m}$ .

To obtain a system of balance equations, the sum of all saturations and the sum of all fluid component concentrations in each phase have to be one:

$$\sum_i S_i = 1 \quad \sum_k c_i^k = 1 \quad i = g, w$$

With respect to the solid phase, the mass balance for the solid phase can be expressed as follows:

$$\phi = \phi_0 - \sum_{k_s}^{r_s} \phi_s^{k_s}$$

where  $\phi_0$  is the reference porosity (initial pore fraction without reactive solid components) and  $\phi_s^{k_s}$  is the pore fraction filled with the solid reactive component  $k_s$ .

Within the source term, the chemical reactions and the operation through the well are integrated into the mass balance. For the fluid components, the source corresponds to:

$$q^k = q_{\text{bio}}^k + q_{\text{geo}}^k + q_{\text{well}}^k$$

Concerning geochemical reactions, the source term for the solid components can be expressed as follows:

$$q^{k_s} = q_{\text{geo}}^{k_s}$$



## 2.1.1 Biochemical reactions

To consider the presence and activity of microorganisms, the interdependent growth and conversion must be modeled. The population kinetics are influenced by the growth and decay of microbes:

$$\frac{\partial(n \cdot S_w \cdot \phi)}{\partial t} = \psi^{\text{growth}} \cdot n \cdot S_w \cdot \phi - \psi^{\text{decay}} \cdot n \cdot S_w \cdot \phi$$

where  $n$  is the microbial density in  $1/\text{m}^3$ ,  $\psi^{\text{growth}}$  is the growth rate in  $1/\text{s}$ , and  $\psi^{\text{decay}}$  is the decay rate in  $1/\text{s}$ .

The growth depends on the quantity of available substrates, as described by the double Monod model:

$$\psi^{\text{growth}} = \psi_{\text{max}}^{\text{growth}} \left( \frac{c_w^{S1}}{\alpha^{S1} + c_w^{S1}} \right) \left( \frac{c_w^{S2}}{\alpha^{S2} + c_w^{S2}} \right)$$

where  $\psi_{\text{max}}^{\text{growth}}$  is the maximum growth rate in  $1/\text{s}$ ,  $c_w^S$  is the mole concentration of substrate  $S$  in the aqueous phase, and  $\alpha^S$  is the half velocity constant of the respective substrate.

To mimic the reduction of the population size due to insufficient substrate supply, the decay is introduced as follows:

$$\psi^{\text{decay}} = b \cdot n$$

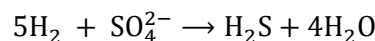
where  $b$  is the decay factor in  $\text{m}^3/\text{s}$ .

Depending on the growth, the reactive components are consumed and produced by the following equation:

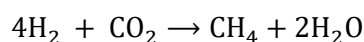
$$q_{\text{bio}}^k = \phi \gamma_{\text{bio}}^k \frac{\psi^{\text{growth}}}{Y} n \cdot S_w$$

where  $\gamma_{\text{bio}}^k$  is the stoichiometric factor and  $Y$  is the yield factor in  $1/\text{mol}$ . Generally speaking, conversion rates increase with increasing growth rates and smaller yield factors.

Within this work, the focus is placed on the biochemical sulfate-reduction and methanation. Sulfate-reducing bacteria are living organisms consuming hydrogen and sulfate ( $\text{SO}_4^{2-}$ ) and simultaneously discharging water and hydrogen sulfide. The stoichiometric equation is as follows:



Methanogenic archaea consume hydrogen and carbon dioxide and produce methane and water. The simplified metabolism reaction takes place according to the Sabatier reaction:



## 2.1.2 Geochemical reactions

Regarding geochemical reactions, two approaches are commonly used: 1) equilibrium models, which are suitable for fast reactions as the reaction is expected to be at equilibrium; 2) kinetic models, where reactions are slower (strong dependency on time) and can therefore be described by a reaction rate [6].

Geochemical effects are considered in the source term in analogy to microbial reactions. However, instead of having a pseudo component that governs the reaction, the reactants in the solid and liquid phases initiate the conversion. In the present study, the implementation of the geochemical reactions is realized with a kinetic model. A generic and straightforward method is a concentration-based model, where the concentrations of the present reactants govern the rate. The following assumes that the reaction is comparatively slow and irreversible, allowing a kinetic implementation similar to [7].

$$q_{\text{geo}}^k = \gamma_{\text{geo}}^k \cdot \kappa \cdot \prod_k^{r_w} \phi \cdot S_w \cdot c_w^k \cdot \prod_{k_s}^{r_s} (1 - \phi) \cdot c_s^{k_s}$$

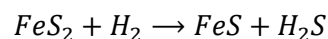
where  $\gamma_{\text{geo}}^k$  is the stoichiometric factor,  $\kappa$  is the reaction rate in  $\text{mol}/(\text{m}^3 \cdot \text{s})$ ,  $r_w$  is the number of reactants in the aqueous phase, and  $r_s$  is the number of reactants in the solid phase.

The impact of the changed porosity on the permeability is modeled by the Kozeny-Carman-relationship.

$$K(\phi) = K_0 \cdot \left( \frac{1 - \phi_0}{1 - \phi} \right)^2 \cdot \left( \frac{\phi}{\phi_0} \right)^3$$

where  $K_0$  is the reference permeability at reference porosity in  $\text{m}^2$ .

For the specific case of pyrite-to-pyrrhotite reduction, the stoichiometric equation can be expressed as follows:



Following the mathematical model, the implementation of the source/sink for the geochemical reaction yields:

$$q_{\text{geo}}^k = \kappa \cdot \gamma_{\text{geo}}^k \phi c_w^{H_2} \cdot S_w \cdot (1 - \phi) \cdot c_s^{FeS_2}$$

## 3 Gitlab repository

The developed model is implemented in DuMu<sup>x</sup> as an additional module and is organized within the Gitlab repository ("dumux-hyuspre") [1]. Gitlab is a development, security, and operation (DevSecOps) environment permitting a collaborative workflow. The provided repository contains relevant instructions for installation, mathematical models, and example cases. For the installation of dumux-hyuspre, the official dumux version 3.6 [8] is required. Regarding example cases, a brief description of the implementation can be found in the following section and the repository. The Gitlab repository is maintained and extended during the project period of HyUSPRe. As Gitlab is a dynamic repository, it is copied into a persistent dataverse [2].

## 4 Simulation cases

The source code composes three test cases:

1. Biogeochemical – test case for the numerical implementation on a simple grid
2. Cornerpoint – test case on a semi-artificial cornerpoint grid with a more complex operation schedule
3. Diffusion – test case to reproduce the diffusion experiments from WP4.

### 4.1 Biogeochemical simulation

The first simulation case allows testing the implemented model of bio-geo-reactive transport during UHS on a simple geometric structure. In the following, the implemented model is briefly explained.

#### 4.1.1 Fluid and solid system

The fluid system comprises two phases (water and gas), which are composed of six components: water (H<sub>2</sub>O), methane (CH<sub>4</sub>), hydrogen (H<sub>2</sub>), carbon dioxide (CO<sub>2</sub>), nitrogen (N<sub>2</sub>), and hydrogen sulfur (H<sub>2</sub>S). Apart from the chemical components, the methanogenic archaea are implemented as an additional pseudo component, which does not affect the fluids' viscosity and density. The characteristic growth parameters are based on recent literature values [9] and are defined as follows:

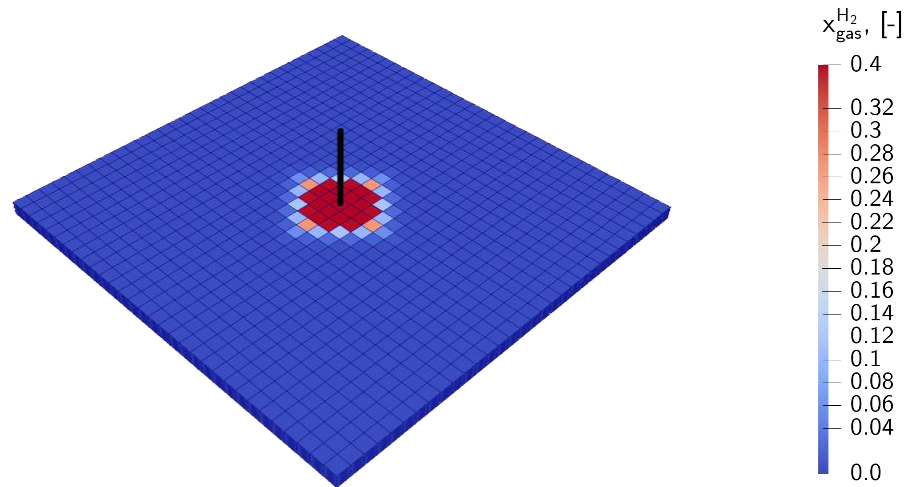
**Table 1: Microbial parameters for the test case**

Parameter	Symbol	Value
Maximal growth rate	$\psi_{\max}^{\text{growth}}$	$1.338 \cdot 10^{-4} \text{1/s}$
H <sub>2</sub> half velocity constant	$\alpha_{H_2}$	$3.6 \cdot 10^{-7}$
CO <sub>2</sub> half velocity constant	$\alpha_{CO_2}$	$1.98 \cdot 10^{-6}$
Yield coefficient	$Y$	$3.9 \cdot 10^{11} \text{1/mol}$
Initial number of microbes	$n^*$	$1 \cdot 10^8 \text{1/m}^3$

Regarding the solid system, three components are introduced: pyrite, pyrrhotite, and one inert component, quartz. In the current state, the geochemical reaction rate is defined artificially.

#### 4.1.2 Spatial discretization

The grid composes a cartesian (YASP) grid with the dimensions 1550m x 1550m x 50m. The discretization is defined with 31x31x10 cells. The distribution of petrophysical properties is homogeneous with  $\phi = 0.2$ ,  $k_x = k_y = 100\text{mD}$ , and  $k_z = 10\text{mD}$ .



**Figure 1: Overview of the spatial domain for the simple simulation case, including the hydrogen mole fraction in the gaseous phase after the injection period**

### 4.1.3 Fluid-matrix interactions

The fluid-matrix interactions regarding advective flux are defined by the model of Brooks & Corey [10] with the following parameters:

**Table 2: Fluid-matrix parameters of the test case**

Parameter	Symbol	Value
Connate water saturation	$S_{wc}$	0.2
Residual gas saturation	$S_{gr}$	0.1
Corey exponent	$\lambda$	2
Capillary entry pressure	$P_{ce}$	0bar

### 4.1.4 Initialization

The initialization is performed based on the hydrostatic equilibrium with  $P_{init} = 100\text{bar}$  at the top of the formation. The saturation is at connate water saturation, and the gas is composed of methane and a minor part of  $\text{H}_2\text{O}$  in thermodynamic equilibrium. The initial volume fraction of pyrite is initially set to 5%. The temperature is set to  $T = 40^\circ\text{C}$ .

### 4.1.5 Operation schedule

A simplified storage operation is defined as 30 days of injection, 30 days idle, 30 days of production, and 15 days idle. The operation is performed rate controlled with identical rates for injection and production ( $1 \cdot 10^6 \text{Sm}^3/\text{d}$ ; 95%  $\text{H}_2$ , 5%  $\text{CO}_2$ ).

## 4.2 Cornerpoint

The second model is a more applicable test case based on a semi-artificial sandstone formation. The test case extends the UHS benchmark scenario [11].

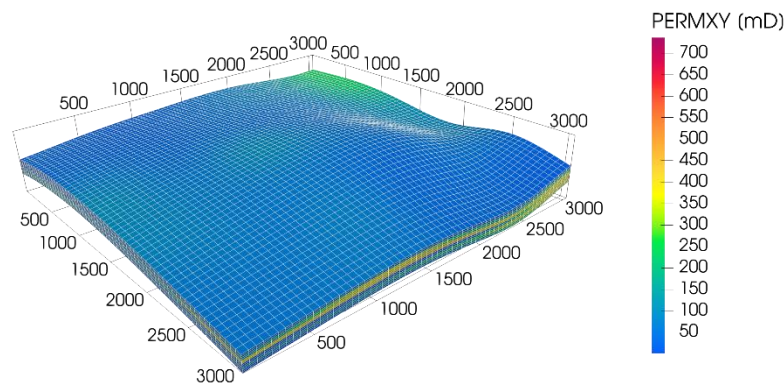
### 4.2.1 Fluid and solid system

A fluid system covering the relevant components is developed. It comprises two phases (water and gas), which are composed of seven components: water ( $\text{H}_2\text{O}$ ), methane ( $\text{CH}_4$ ), hydrogen

(H<sub>2</sub>), carbon dioxide (CO<sub>2</sub>), nitrogen (N<sub>2</sub>), hydrogen sulfur (H<sub>2</sub>S), and sulfate (SO<sub>4</sub><sup>2-</sup>). Apart from the chemical components, two types (sulfate-reducing and methanogenic) of microorganisms are implemented as additional pseudo components, which do not affect the fluid viscosity and density. The characteristic growth parameters are based on literature values ([9], [12]). Regarding the solid system, three components are introduced: pyrite, pyrrhotite, and as an inert component, quartz. The rate for the geochemical reaction is currently artificial.

## 4.2.2 Spatial discretization

A corner-point grid based on a semi-artificial geological structure (cf. Figure 2) is used for the spatial discretization of the simulation. It consists of 44652 (61x61x12) grid cells with a dimension of 50 m x 50 m (x- and y-direction) and a varying thickness. The petrophysical properties (porosity and permeability) are distributed heterogeneously, and the permeability is additionally defined as anisotropic. The average porosity is 15%, and the mean horizontal permeability is 143mD ( $k_v \sim 3\text{mD}$ ), which can be observed in some sandstone formations in Northern Germany (permeability distribution based on a modified poro-perm-correlation).



**Figure 2: General overview of the geological structure (here: horizontal permeability)**

## 4.2.3 Initialization

The system is initialized with a pressure of  $P_{GWC} = 81.6\text{bar}$  at the gas-water-contact, which is located at a depth of 1210m. A transition zone is established by the capillary pressure (Brooks-Corey parameter  $\lambda = 2.0$ ,  $P_e = 0.1\text{bar}$ ) separating the gas and water zone. The initial gas composition in the gas zone is defined as typical natural gas. Furthermore, the initial pyrite concentration is set to 0.1%, and the sulfate content in the liquid phase is 0.0003375.

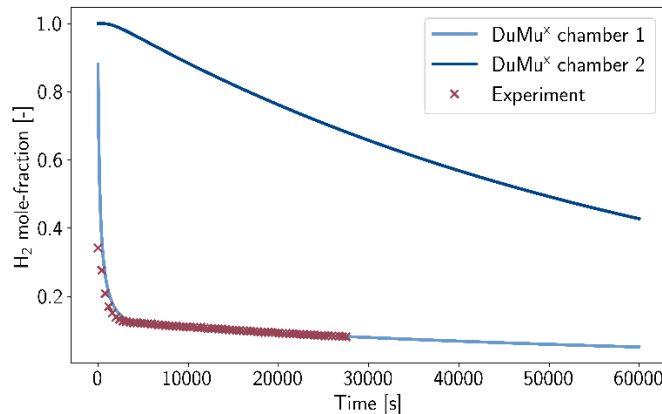
## 4.2.4 Operation schedule

In the present study, the schedule comprises two sections: 1) Conversion from natural gas storage into UHS and 2) Regular storage operation. In both sections, the injection/production occurs along a single well located in the centre of the structure. The injected gas composition remains constant (10% H<sub>2</sub>, 90% natural gas). The first section, the conversion cycle, is characterized by a bottom-hole pressure controlled injection which is incrementally increased from 90bar to 102bar (step size: 4bar) to increase the reservoir pressure and raise the hydrogen content in the storage. Four cycles of 60 days with one month idle time between each cycle are performed. After the conversion cycles, regular storage cycles are conducted. The regular operation consists of alternating injection and production with a constant rate of  $q = 293.13 \text{ mol/s} = 6 \cdot 10^5 \text{ Sm}^3/\text{day}$ . The duration of injection is identical to the withdrawal

(90 days) to equalize the cumulative volumes. Like the conversion cycles, the regular storage cycles are separated by idle times (30 days).

### 4.3 Diffusion

The last case allows the reproduction of the diffusion measurements performed within the scope of WP4 [13].



**Figure 3: Comparison of hydrogen fractions versus time observed in the laboratory and modeled in DuMu<sup>x</sup> for the reference sample at reference conditions (T=40°C and p=100bar)**

#### 4.3.1 Modeling of binary diffusion coefficients

Within HyUSPRe, more than 30 experiments of molecular diffusion were conducted to characterize the mixing of hydrogen and methane within the gaseous phase. Based on these measurements, correlations for the effective binary diffusion coefficients were developed as a function of thermodynamic and petrophysical properties. The general form for the effective binary diffusion can be expressed as follows:

$$D_{pm}^{AB} = \phi \cdot \tau \cdot S_g \cdot D^{AB}$$

The developed model consists of two correlations:

1. Tortuosity as a function of porosity, permeability, and saturation considering the presence of the porous medium:

$$\tau = \phi \cdot S_g \cdot k_{\text{eff}}^{\frac{1}{5}} \cdot 176.916 \text{ m}^{-\frac{2}{5}}$$

2. Bulk diffusion coefficient as a function of pressure and temperature:

$$D^{AB}(T, p) = \beta_1 + \beta_2 \cdot T + \beta_3 \cdot p + \beta_4 \cdot T^2 + \beta_5 \cdot T \cdot p + \beta_6 \cdot p^2$$

$$\begin{aligned} \beta_1 &= 3.61069 \cdot 10^{-5} \text{ m}^2/\text{s} & \beta_4 &= 1.67793 \cdot 10^{-10} \text{ m}^2/(\text{s} \cdot \text{K}^2) \\ \beta_2 &= -1.46672 \cdot 10^{-7} \text{ m}^2/(\text{s} \cdot \text{K}) & \beta_5 &= 2.95155 \cdot 10^{-15} \text{ m}^2/(\text{s} \cdot \text{K} \cdot \text{Pa}) \\ \beta_3 &= -1.74842 \cdot 10^{-12} \text{ m}^2/(\text{s} \cdot \text{Pa}) & \beta_6 &= 3.71863 \cdot 10^{-20} \text{ m}^2/(\text{s} \cdot \text{Pa}^2) \end{aligned}$$

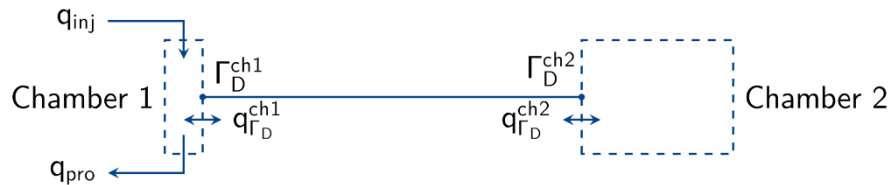
Further information regarding the evaluation of the experiments and the consecutive development of the correlations can be found in [13].

### 4.3.2 Fluidsystem

The fluid system is composed of two phases (liquid and gases) with seven components ( $H_2O$ ,  $CH_4$ ,  $H_2$ ,  $CO_2$ ,  $N_2$ ,  $H_2S$ , and  $SO_4^{2-}$ ) developed to investigate underground hydrogen storage scenarios.

### 4.3.3 Spatial discretization

For the spatial discretization, representing the core sample ( $\approx 6cm$ ) between the two chambers, a one-dimensional grid with 100 equidistant elements is defined. While the first chamber possesses a relatively large volume, the second represents only the gas distribution element. An extrusion factor corresponding to its area is introduced to consider the axial cross-section of the rock specimen. The box method (control volume finite element method) is selected for the discretization method due to its versatile possibilities of evaluating gradients locally. The porosity and permeability of the domain are defined homogeneously and parameterized with the measured values from the laboratory.



**Figure 4: Schematic representation of the domain, including the chambers' modeling.**

### 4.3.4 Initialization and boundary conditions

For the initialization of the system, all elements contain only gas, which is composed entirely of hydrogen, following the experimental procedure. The pressure and temperature are defined as the experimental conditions. To model the two chambers at the sides of the core sample (cf. Figure 4), time-dependent Dirichlet boundaries are used to mimic the changing concentrations of hydrogen and methane within the chambers.

**Table 3: Overview of the definition of the primary variables of the Dirichlet boundaries for chamber 1 and 2**

Pri.variables	Chamber 1	Chamber 2
$p_g$	$p_{init} = p_{exp}$	$p_{init} = p_{exp}$
$S_w$	0	0
$c_g^k$	$c_{ch1,t}^k = \frac{n_{ch1,t-1}^k - (q_{\Gamma_D}^k + q_{inj}^k - q_{pro}^k)\Delta t}{\sum_k n_{ch1,t-1}^k - (q_{\Gamma_D}^k + q_{inj}^k - q_{pro}^k)\Delta t}$	$c_{ch2,t}^k = \frac{n_{ch2,t-1}^k - q_{\Gamma_D}^k \Delta t}{\sum_k n_{ch1,t-1}^k - q_{\Gamma_D}^k \Delta t}$

---

## 5 Concluding remarks

Using numerical reservoir simulation as an accompanying tool during the operation of underground energy storage possesses a high potential to optimize the overall storage process. As most reservoir simulators focus on modeling the fluid transport of hydrocarbons, these simulators may be insufficient to model the unique process of UHS, such as bio- and geochemical reactions.

This study extends the open-source simulator DuMu<sup>x</sup> by an additional module to provide a proper framework for simulating UHS-related processes. Here, the focus is placed on bio- and geochemical effects and the gas-gas mixing by molecular diffusion enriched with data from laboratory observations. In the current state, the module contains three test scenarios: 1) biogeochemical - a simple case testing the overall flow process; 2) cornerpoint – a more complex case with the interface for typical reservoir simulators (.GRECL) for simplified inread of complex grid structures; 3) diffusion – test case to reproduce the results of the one-chamber diffusion measurements.

The Gitlab repository is regularly updated and maintained during the project period of HyUSPRe, and further extensions are planned.



---

## References

- [1] S. Hogeweg, "GitLab Repository - dumux-hyuspre." Apr. 11, 2023. [Online]. Available: <https://gitlab.tu-clausthal.de/asho13/dumux-hyuspre/>
- [2] S. Hogeweg and B. Hagemann, "Data for: HyUSPRe - Work Package 6 - Validated open-source reservoir modelling software that can simulate coupled flow, geochemical and microbiological processes in a porous reservoir under operational hydrogen storage conditions." GRO.data, 2023. doi: 10.25625/GNHQ93.
- [3] B. Flemisch *et al.*, "DuMux: DUNE for Multi-(Phase, Component, Scale, Physics,...) Flow and Transport in Porous Media," *Adv. Water Resour.*, vol. 34, no. 9, pp. 1102–1112, 2011, doi: 10.1016/j.advwatres.2011.03.007.
- [4] T. Koch *et al.*, "DuMux 3 – An Open-Source Simulator for Solving Flow and Transport Problems in Porous Media with a Focus on Model Coupling," *Comput. Math. Appl.*, 2020, doi: 10.1016/j.camwa.2020.02.012.
- [5] B. Hagemann, *Numerical and Analytical Modeling of Gas Mixing and Bio-Reactive Transport During Underground Hydrogen Storage*, 1. Auflage., vol. Band 50. in Schriftenreihe des Energie-Forschungszentrums Niedersachsen, vol. Band 50. Göttingen: Cuvillier Verlag, 2018. [Online]. Available: <https://ebookcentral.proquest.com/lib/gbv/detail.action?docID=5218530>
- [6] N. Hassannayebi, S. Azizmohammadi, M. Lucia, and H. Ott, "Underground hydrogen storage: application of geochemical modelling in a case study in the Molasse Basin, Upper Austria," *Environ. Earth Sci.*, vol. 78, no. 5, p. 177, 2019, doi: 10.1007/s12665-019-8184-5.
- [7] A. S. Abd and A. S. Abushaikha, "Reactive transport in porous media: a review of recent mathematical efforts in modeling geochemical reactions in petroleum subsurface reservoirs," *SN Appl. Sci.*, vol. 3, no. 4, p. 401, 2021, doi: 10.1007/s42452-021-04396-9.
- [8] Y. Wang *et al.*, "GitLab Repository - dumux 3.6.0." Dec. 16, 2022. [Online]. Available: <https://git.iws.uni-stuttgart.de/dumux-repositories/dumux/-/releases/3.6.0>
- [9] G. Strobel, B. Hagemann, M. Wirth, and L. Ganzer, "Pore-Scale Modeling of Microbial Growth in A Two-Phase Saturated Porous Medium," vol. 2020. European Association of Geoscientists & Engineers, pp. 1–14, 2020. doi: 10.3997/2214-4609.202035171.
- [10] R. H. Brooks and A. T. Corey, "Hydraulic Properties of Porous Media and Their Relation to Drainage Design," *Trans. ASAE*, vol. 7, no. 1, pp. 0026–0028, 1964, doi: 10.13031/2013.40684.
- [11] S. Hogeweg, G. Strobel, and B. Hagemann, "Benchmark study for the simulation of Underground Hydrogen Storage operations," *Comput. Geosci.*, vol. 26, no. 6, pp. 1367–1378, Dec. 2022, doi: 10.1007/s10596-022-10163-5.
- [12] B. Hagemann, L. Ganzer, and M. Panfilov, "Field Scale Modeling Of Bio-Reactions During Underground Hydrogen Storage," presented at the ECMOR XVI - 16th European Conference on the Mathematics of Oil Recovery, in Proceedings. Barcelona, Spain: EAGE Publications BV/Netherlands, 2018. doi: 10.3997/2214-4609.201802116.
- [13] J. Michelsen *et al.*, "Hydrogen reservoir flow behaviour: Measurements of molecular diffusion, mechanical dispersion and relative permeability," H2020 HyUSPRe project report, Apr. 2023.