In-situ Measurement of gas distribution generated by continuous and pulsed gas injection

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Eine gezielte Steuerung der Gasausbreitung bei einer in-situ Injektion mit Sauerstoff, Luft, Methan oder Kohlendioxid in Grundwasser entscheidet über die Raumwirkung der Gasinjektion, über die Effizienz der Gasspeicherung und über die Einlösungskinetik der Gasphase in das kontaminierte Grundwasser. Gegenstand der Studie sind die eigenen Arbeiten zur Direktgasinjektion von Luft an einem konkreten Versuchsfeld auf der Feldskala. Das Versuchsfeld befindet sich am Chemiestandort Leuna. Der Standort ist Teil eines ausgedehnten Schadensbereiches auf dem Gelände der Alten Raffinerie. Der experimentelle Nachweis von Gasströmungsmechanismen erfolgt im Zuge systematischer Gasinjektionsexperimente mit Variationen von Injektionsraten und Impulsbreiten unter Anwendung von gezielter Niedrig- und Hochdruckgasinjektion. Die dabei erzielten Raumwirkungen und Sättigungsverteilungen wurden durch Nutzung eines 3D-Sensorarrays eingehend ausgewertet. Die Gasinjektionsszenarien für den Feldstandort wurden mittels numerischer Modellierung mit TOUGH2 (PRUESS, 1991) verifiziert und untersucht. Unsere Feldexperimente zeigen sehr deutlich, dass die resultierenden Gasströmungsmuster stark von der gewählten Injektionsrate abhängen. Vergleiche unserer Feldexperimente mit den Simulationsergebnissen erbrachten, dass numerische Simulatoren wie TOUGHII, welche auf Kontinuumsansätzen (Darcy-Gesetz) basieren, nicht in der Lage sind, inkohärenten Gasblasentransport zu reproduzieren.

Gas flow regime, for example in oxygen, air, methane, and carbon dioxide injection, highly affects the mass transfer, degradation and reaction phenomenon, and changes the efficiency of gas injection process. In this study we injected air under different injection scenarios at field site Leuna. Leunawerke is a chemical plant in Germany polluted by chemical compounds from nearby industrial site. In each scenario we injected gas continuously, in short pulses, or in combination of both scenarios and measured the gas saturation in three dimensions by a high resolution network of gas sensors. We simulated gas injection scenarios with TOUGH2 numerical model which is implemented by Karsten Pruess in 1991. Our experimental investigations in field site showed that gas flows pattern highly depends on gas injection rate. Comparison between our experimental and simulation results revealed that numerical simulator, like TOUGH2, which assumed continuous fluid flow equations (e.g. Darcy Law) can not model a regime where gas flows in separated bubbles.

1 Introduction

Different gas like oxygen, air, methane, carbon dioxide has been injected in soil for different purposes in last two decades. For example air injected to remove efficiently the volatile organic compounds (VOCs e.g. industrial fuels and solvents) from contaminated ground water

(JI et al. 1993, SEMER et al. 1998, MCCRAY 2000, THOMSON & JOHNSON 2000, REDDY et al. 2001).

Determining the gas flow pattern through the porous media is important in gas injection because gas flow pattern highly affects the mass transfer, degradation and reaction phenomenon, and changes the efficiency of process. Many studies have been done to investigate the gas flow pattern in laboratory (JI et al. 1993, MCCRAY & FALTA 1997, SEMER et al. 1998, REDDY & ADAMS 2001, SELKER et al 2007, GEISTLINGER et al. 2009, STAUFFER et al. 2009). In most laboratory experiments, glass beads were used instead of irregular grains of soil, but water remained with same specification on wetting the surface. JI et al. (1993) visualized flow patterns in 2 dimensions (2D) and related the flow regime to the grain size. THOMSON and JOHNSON (2000) provided an overview of mathematical modeling efforts and emphasized that the air entry pressure governs migration pattern at grain scale, while at the field scale low permeability zone alters the bulk flow. But THOMSON and JOHNSON (2000) they did not perform any field experiment to verify their argument.

REDDY and ADAMS (2001) declared that injected air in homogeneous coarse sand travels in channels (channelized flow or coherent flow) whereas it flows in bubbles (bubbly flow or incoherent flow) in gravel profiles. They stated channelized flow creates a parabolic zone and has a bigger zone of influence.

GEISTLINGER et al (2006) investigated direct gas injection in 2D laboratory experiments. In spite of JI et al. (1993) they found that transition from coherent to incoherent flow depends on both grain size and flow rate. STAUFFER et al. (2009) performed 2D direct gas injection in laboratory and applied a new method for cell size in numerical simulation. However this method simulated the results better than macro-scale modeling but it could not show the channelized air flow observed in experiments.

Unfortunately glass beads which were used in laboratory experiments can not reflect the soil characteristics. On the other hand previous laboratory experiments were performed in 2D. Hence we performed a comprehensive direct gas injection in field site Leuna to investigate the gas flow in 3D through the soil profile. Leuna site locates near to Leunawerke and has been polluted by chemical compounds from nearby industrial site. We developed a network of gas sensors and measuring devices in Leuna and performed different air injection scenarios. In each scenario gas injected continuously or in short pulses with specific injection rate. We monitored the gas saturation in three dimensions by in-situ sensors and used TOUGH2 program to simulate the injection process. TOUGH2 is a numerical simulation program which is implemented by KARSTEN PRUESS (1999) and has been used to model the fluid and heat flows.

Our experimental investigations in field site showed bubbly flow in low pressure injection (injection rate less than 0.2 (m^3/hr)) and channelized flow in high pressure gas injection (injection rate bigger than 1.5 (m^3/hr)). Comparison between results of experiments and simulation revealed that numerical simulator like TOUGH2 which has used continuous fluid flow equations (e.g. Darcy Law) can not model the bubbly flow. On the other hand natural sedimentation caused the vertical permeability becomes smaller (usually 5 to 10 times) than horizontal one. We applied distinct permeability values in vertical directions in numerical simulation to represent this natural anisotropy.

2 Field Experiment

Working hypotheses for field application is derived from laboratory experiments done by GEISTLINGER et al. (2006). They investigated the transition from coherent to incoherent flow in 2D tanks filled with glass beads and found that transition depends on both grain size and flow rate. GEISTLINGER et al. (2006) stated that very low flow rates results in incoherent flow which establishes high gas saturation in a certain region, more than coherent flow.

Gas injection in Leuna was a smart development of high pressure injection (HPI) and low pressure injection (LPI) scenarios to to investigate the statements of GEISTLINGER et al. (2006) and study the effect of injection rate on flow pattern in soil texture.

2.1 Sensor Array Development and Injection Scenarios

We did the remediation experiments within the 20 m^2 area which is called sensor field. We installed 5 capacitive moisture sensors (Sensatec, GmbH, Germany) , 16 redox-potential sensors (Sensatec, GmbH, Germany), 16 temperature sensors (Sensatec, GmbH, Germany), and 55 oxygen sensors (Presens GmbH, Germany) in sensor field. Fig. 1 shows the arrangement of injection well, oxygen sensors, and observation wells in sensor field.

We used four injection scenarios which are subjected to the following conditions: (1) gas injected through GP11. (2) Three cubic meter of gas injected during the performance of each test. (3) Gas saturation values were measured during the injection, at the end of injection and after the injection process.

Table. 1 shows characterizations of injection scenarios where two first experiments refer to low pressure injection (LPI), third one is high pressure injection (HPI), and the last one is a combination of LPI and HPI processes.

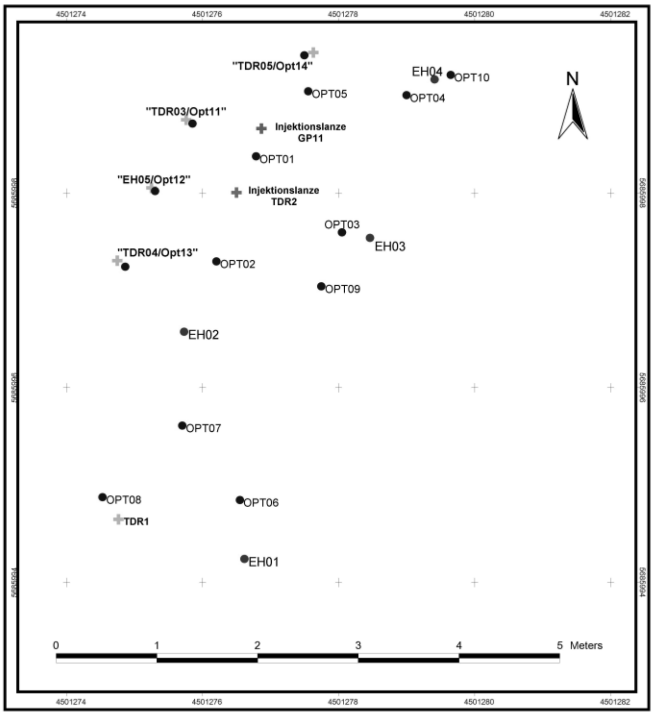

Fig. 1: Arrangement of injection and observation wells in sensor field

Table. 1: Characterization of injection scenarios

Scenario	*Injection rate (m^3/hr)*	*Injection pressure (bar)*	*Time of gas injection (hr)*	*Time of monitoring (weeks)*	*Volume of injected gas (m^3)*
E 1.1	1.5	1.9	2	2	3
E 2.1	0.18	0.9	16.6	2	3
E 3.1	2.4	8	1.25	3	3
E 4.1	2.4	5.5	0.7	-	1.68
	0.18	0.9	7.33	2	1.32

3 Numerical Modeling Using TOUGH2

TOUGH2 is an open source program to simulate heat and fluid flows in porous and fractured media. TOUGH2 code provided different units of fluid property or EOS (equation-of-state). We used EOS3 which describes gas-water flow under isothermal conditions.

3.1 Parameterization of Simulation Model

Measurement in sensor field shows that two layers exist at top and bottom of aquifer which have low permeability. As the low-permeable layer at top is not right above the aquifer, the saturated part should also be considered in simulation. Increasing the size of model demands higher CPU usage and increases the simulation time. So we considered a smaller size for saturated part.

The simulation model extended 6.8 meter in each X, Y, and Z directions where the single cell dimension was Lx.Ly.Lz=20x20x20 cm^3. GP11 is located at the center of model and perforated 20 cm at the bottom, right above the low-permeable layer. Before injection process, saturation exhibits inhomogeneous transition from complete water saturation at the bottom to complete air saturation at the top. We considered Dirichlet boundary conditions (constant properties) at the top and bottom of the model and run TOUGH2 to achieve the inhomogeneous condition by applying the capillary-gravity equilibrium. Equilibrium distribution is used as initial condition for injection part. Boundary condition for injection process is considered infinite acting in all direction similar to field site. Fig. 2 shows the top view of simulation discritization respect to sensor field site in Leuna. Soil properties for simulation are estimated by geological profile and sieve analysis data.

Fig. 3 shows different rock types between 3 m and 9.7 m depth in OXY09 which is located in sensor field. We used the geological information in this location to estimate soil properties, especially in vertical direction.

Table. 2 shows the mean values of soil properties e.g. porosity. Mean absolute permeability in Table. 2 is changed in simulation corresponding to heterogeneity in horizontal and vertical directions.

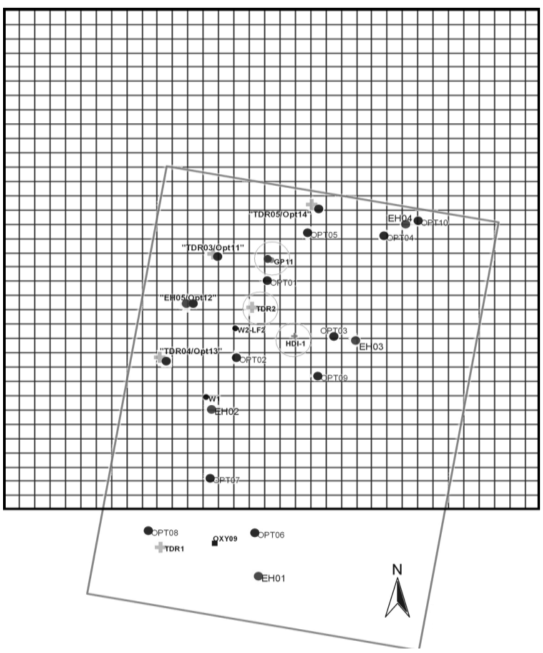

Fig. 2: Top view of simulation discritization respect to injection site

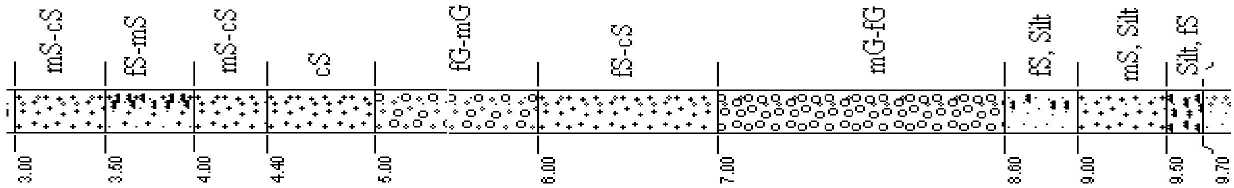

Fig. 3 : Geological profile in OXY09; Capital letters are rock type and small letters refer the grain size

Table. 2: Characterization of injection scenarios

Parameters	*Value*
Porosity (%)	30
Grain density (kg/m^3)	2650
K $_{absolute}$ homogeneous porous media (m^2)	1.17E-11
K $_{absolute}$ low-permeable layer at top and bottom (m^2)	1.00E-18

3.2 Heterogeneity in Horizontal and Vertical Directions

Hydraulic properties like permeability highly depend on pore size and connectivity between pores. Size of pore radius is varying through any porous media even in a homogeneous one, like a pack of pure equal-size glass beads. Natural sedimentation has created differences in pore size and hydraulic properties through any soil texture. Hence we should use a distribution of soil properties to apply this kind of variation in horizontal and vertical direction.

Injection log characterize hydraulic conductivity especially in horizontal direction. We used data of injection logs and geological profile to create a stochastic permeability field by the use of SGSIM program (Sequential Gaussian simulation program, DEUTSCH & JOURNEL, 1998). A stochastic field is a set of random variables defined over a field of interest according to filed specification like mean value of data.

Groundwater Modeling System (GMS, developed by Aquaveo, LLC in Provo, Utah) is a comprehensive graphical user environment for performing groundwater simulations. We input the permeability values of injection logs and geological profile to GMS and GMS gave back the specification values of permeability field like mean and deviation of values about the mean. We used these specification values to create a stochastic permeability field by SGSIM program.

We have the permeability values in location OXY01, OXY03, OXY05, OXY07, OXY09, OXY012, GP03, GP05, GP09 and GP11.SGSIM takes these values as reference numbers to statistically simulate the permeability values in other location. Actually SGSIM does not simulate the permeability values at reference points. SGSIM needs some specification like type of grid search and parameters like search radius. We chose 3.7 and 0.37 cm for search radius in horizontal and vertical direction respectively. Because the vertical permeability is smaller (5 to 10 times) than horizontal one. Results of SGSIM are the values of stochastic permeability field which should be divided by mean permeability value to give us the permeability modifiers. Values of permeability modifiers are entered in MESH file (position 41-50) and multiplied by absolute permeability by TOUGH2 simulation. In order to imply the modifier, SEED option should be activated in TOUGH2 simulation by writing the SEED in ROCK block in INPUT file.

Simulation of Bench scale experiments (GEISTLINGER et al. 2009) reveals that the stochastic variability in porosity and permeability field could not explain the experimental results. Instead, employing capillary pressure heterogeneity could explain the saturation patterns observed in the laboratory experiments. When SEED option is activated, TOUGH2 uses Leveret scaling to adjust capillary pressure respect to the permeability variation.

Different realizations for permeability field which are created by SGSIM have the same chance to happen in reality, so they are called equiprobable (i.e. have the same probability). Fig. 4 shows two equiprobable realizations by SGSIM where desired data for sensor field should be cut out. Left picture in Fig. 4 is Realization1 (R1) and right one is Realization2 (R2). We create a stochastic field bigger than sensor field because we can use more data as input data to GMS and it gives possibility to simulate the injection process from location out of the sensor field. Interpreting the measured gas saturation during the injection process in sensor field revealed a low permeability zone in North part of sensor field. So among all realizations we chose ones which has lower permeability distribution in north part of sensor field.

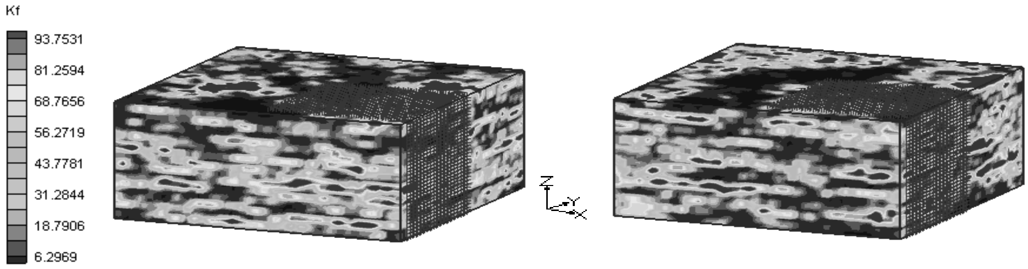

Fig. 4 : Different realization for stochastic horizontal permeability filed

Most investigations like core analysis and Injection logs have revealed that vertical permeability is smaller than horizontal one. However vertical permeability is usually considered ten times smaller than horizontal one but there is no strict relation between these two permeabilities. Some measurement of soil properties in vertical direction like electrical conductivity (EC) shows big variation in rock characterization in vertical direction. Fig. 5 is an example of high variation in EC-value in vertical direction at GP11 in sensor field.

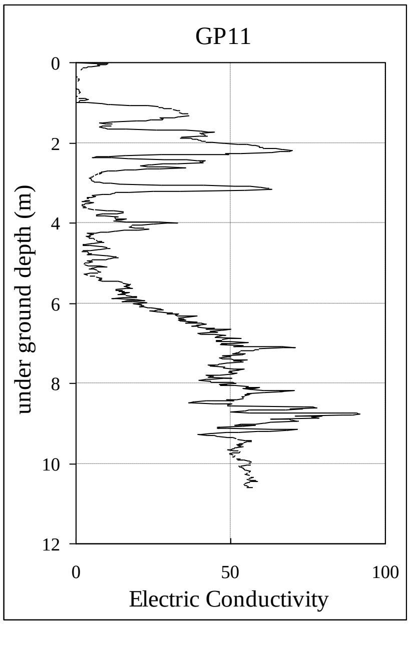

Fig. 5 : Measurement of electrical conductivity in vertical direction at GP11

On the other hand SGSIM uses Gaussian distribution function to create the permeability field and creates a continuous field. So we applied distinct permeability values vertical direction to represent natural anisotropy between permeability in two directions. We estimated the position of these layers from geological profile and experimental gas saturation measurement in TDR2.

The results of gas saturation measurement during the injection process can give some valuable hints about the location of low and high permeable zones. The hypothesis in this study is that gas flows in pores which have low resistance to flow and the region with high accumulation of gas indicates a low-permeable region in upper region. We defined different rock types to identify the layers in vertical direction. Corresponding rock type of each grid comes in MESH file (position 20). Values of absolute permeability in all directions (x, y, and z) for each rock type are specified in INPUT file.

We used layering classification to show the effect of varying permeability in vertical directions, but heterogeneity in vertical direction does not distribute in layers. Permeability in vertical direction has a stochastic distribution and it needs to use some software like Transition Probability Geostatistical Software (TPROGS). In spite of SGSIM, TPROGS can create the stochastic fields with distinct classification of rock types.

4 Results and Discussion

4.1 Gas Flow Pattern

Measurement of gas saturation in field experiments revealed that transition from coherent to incoherent flow through a specific porous media can be controlled by injection pressure (or injection rate). In injection process, gas pressure has to be increased enough to overcome the capillary pressure and invade a pore. When the gas pressure is not enough to invade the next pores, gas can not flow as a channel and it moves as separated bubbles. The separated bubbles tended to move upward with a parabolic spreading. Incoherent flow has been created by low pressure gas injection (LPI). Bubbly flow results in higher gas saturation through the porous media. Coherent gas flow indicates gas flow driven by an applied pressure gradient along a continuous gas phase. High pressure gas injection (HPI) creates coherent channelized flow where viscous force over comes the capillary pressure. In channelized flow gas spreads through the porous media more horizontally.

Fig. 6 shows measured gas saturation in location TDR2 by moisture sensor for injection scenarios mentioned in Table. 1. In Fig. 6 values of gas saturation are reported during the injection, at the end of injection and after the injection process which corresponding time is mentioned in the legend. Fig. 6 shows the increasing of gas saturation in top layers in all scenarios after stopping the injection process which is an evidence of gas movement because of gravity force. Negligible reduction in gas saturation 2 hours after stopping the injection in E 2.1 is a strong evidence of incoherent gas flow in Fig. 6. Change in gas saturation in incoherent flow is not obvious because gas bubbles accumulate as long as storage capacity of the invaded region is reached. But for coherent flow one expects noticeable changes in gas saturation after stopping injection that shows the gas movement. Laboratory investigation by LAZIK et al. (2008) has proved these evidences. LAZIK et al. (2008) observed a significant difference between coherent moving gas and discontinuous residual gas which is formed behind the front after stopping the injection.

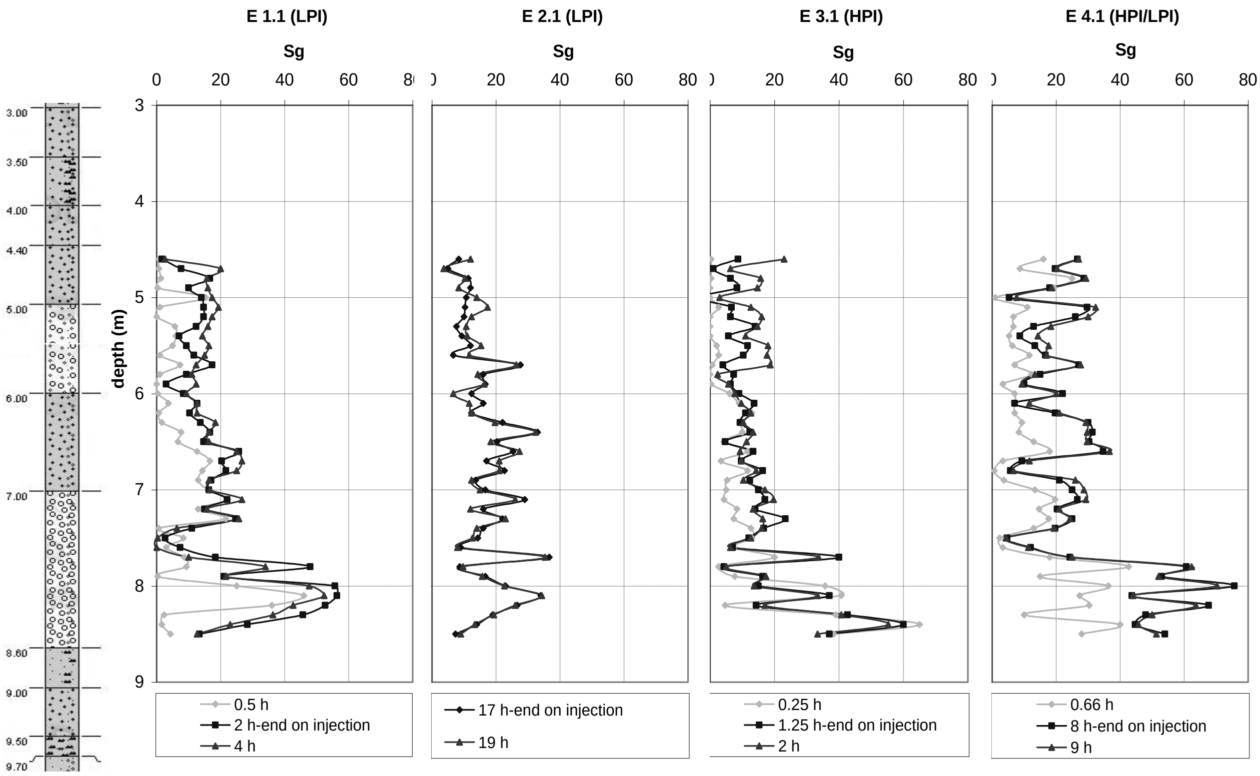

Fig. 6 : Gas measurement at location TDR2 for injection scenarios of Table. 1

The obvious differences between gas saturation profile in Fig. 6 show that each scenario creates specific gas flow pattern. We repeated injection process E 1.1 as a low pressure injection and E 3.1 as a high pressure injection processes and called them E 1.2 and E 3.2 which emphasize repetition by 2. Fig. 7 shows that gas saturation in E 1.2 and E 3.2 have the same trend of maximum and minimum values as E 1.1 and E 3.1 respectively and confirmed that gas pattern depends on injection pressure. The pattern which has been created under specific condition allows the gas passes easily in repetition processes and caused the measured gas saturation in repetition processes becomes smaller.

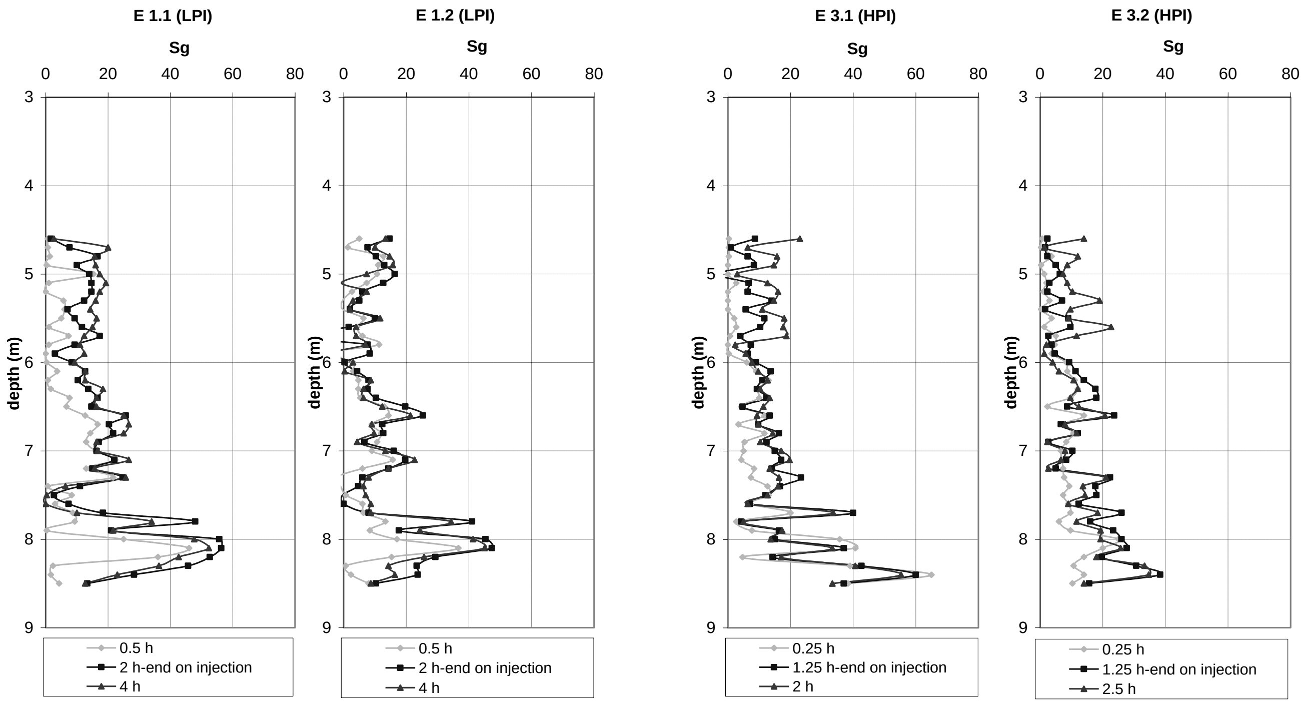

Fig. 7 : Gas measurement at location TDR2 for repetition of injection scenarios E 1.1 and E 3.1

4.2 Simulation Results

Fig. 8 shows the gas saturation profile at TDR2 for injection scenarios E 1.1 (Table. 1) respect to experimental profile. Simulation results in Fig. 8 are obtained when the heterogeneity in permeability and capillary pressure are applied in TOUGH2 simulation. But the effect of distinct permeability in vertical direction has been shown in one saturation profile. In this work we applied distinct permeability in vertical direction by considering the low and high permeable layers. The simulation results when layering is considered shows better results respect to the one in which the layering effect is not considered. Differences between simulation results in Fig. 8 show the effect of considering heterogeneity in vertical direction rather. The time and rate of each simulation is mentioned in the Fig. 8.

GMS has different modules to visualize the gas flow pattern, especially in 3D, so we used GMS to visualize the simulation results. Fig. 9 shows the gas saturation in 3D for simulation results which are presented in Fig. 8. Right and left gas distributions in Fig. 9 show gas saturation in E 3.1 scenario at the end of injection and in E 1.1 during the injection.

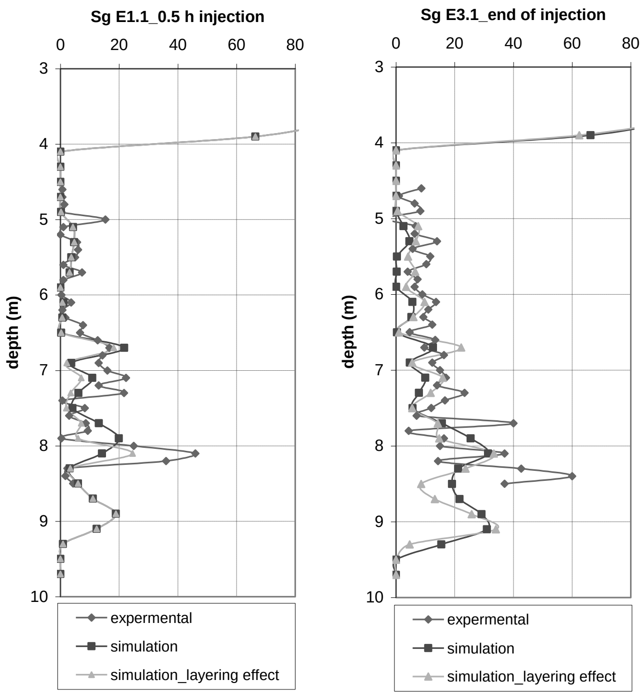

Fig. 8 : Comparison between simulation results and experimental measurement

The gas distribution in Fig. 9 shows how the injection pressure can affect the flow and expansion of gas through the porous media. However TOUGH2 can simulate the gas saturation but this program is not able to simulate bubbly and channelized flow. In other words there is no difference exist between the visualized gas saturation of LPI with bubbles and HPI with channels in Fig. 9.

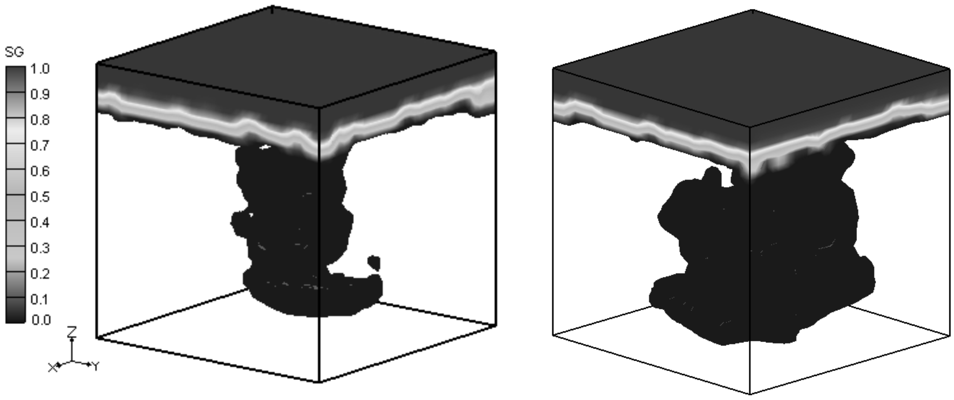

Fig. 9 : Visualization of simulated gas saturations in 3D

5 Conclusion

Analyzing the experimental gas saturations measured by moisture sensors showed the gas flow pattern depends on gas injection rate (or pressure). Our investigations in field site showed that LPI (e.g. injection rate less than 0.2 (m^3/hr)) creates bubbly flow and HPI (e.g. injection rate higher than 1.5 (m^3/hr)) builds gas channels which let the gas goes upward faster than bubbly flow. On the other hand bubbly flow creates a region with high gas saturation. So for accumulating the gas in a region, LPI scenario should be used.

Gas distribution in Fig. 9 showed that gas expands wider in horizontal direction in HPI scenario like E 3.1 respect to LPI scenario like E 1.1. This conclusion confirmed the Geistlinger et al. (2009) observation in their optical bench-scale experiments. Geistlinger et al. (2009) observed that gas plum expands in horizontal direction when gas injected with high flow rate. So HPI should be used when the main purpose of gas injection is transferring the gas in a long distance.

Applying the continuous heterogeneity field created by SGSIM can not represent the discontinuous heterogeneity in vertical direction. Simulation results showed that the heterogeneity in vertical direction should be applied rather than heterogeneity in horizontal direction. In this work we implied the layered permeability in vertical direction. Fig. 8 showed how applying the distinct permeability in vertical could change the simulation results. But heterogeneity in vertical is a stochastic field like heterogeneity in horizontal direction. However we applied layered permeability but a distinct stochastic permeability field in vertical direction should be applied in next simulation study.

Comparison between our experimental and simulation results revealed that numerical simulator which assumed continuous fluid flow equations (e.g. Darcy Law) can not model the bubbly flow. TOUGH2 uses Darcy law for gas and liquid flows and it can not represented the separated bubbles in low pressure gas injection. In Fig. 9 there is no big difference between the gas plums of E 1.1 and E 3.1, however E 1.1 is LPI and E 3.1 is HPI. Simulated gas saturation profile for LPI scenarios (especially for E 2.1) deviated by 30% from the experimental measurement. Deviation of gas saturation in LPI is also confirms that TOUGH2 is not able to simulate the bubbly flow. The open question raised by the present study is: which flow equation should be used in simulation job and how we can apply these flow equations in simulation job to model the bubbly flow?

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