Available online at www.sciencedirect.com

ScienceDirect

Energy Procedia 40 (2013) 25 – 33

Energy Procedia

European Geosciences Union General Assembly 2013, EGU

Division Energy, Resources & the Environment, ERE

Research sites of the H2STORE project and the relevance of lithological variations for hydrogen storage at depths

Henkel, S.[a]*, Pudlo, D.[a], Gaupp, R.[a] and the H2STORE-Team

[a]*Friedrich Schiller University Jena, Institute of Geoscience, Burgweg 11, D-07749 Jena, Germany*

Abstract

The H2STORE collaborative project investigates potential geohydraulic, petrophysical, mineralogical, microbiological and geochemical interactions induced by the injection of hydrogen into depleted gas reservoirs and CO_2- and town gas storage sites. In this context the University of Jena performs mineralogical and geochemical investigations on reservoir and cap rocks to evaluate the relevance of preferential sedimentological features, which will control fluid (hydrogen) pathways, thus provoking fluid-rock interactions and related variations in porosity and permeability. Thereby reservoir sand- and sealing mudstones of different composition, sampled from distinct depths (different: pressure/temperature conditions) of five German locations are analysed. In combination with laboratory experiments the results will enable the characterization of specific mineral reactions at different physico-chemical conditions and geological settings.

Selection and peer-review under responsibility of the GFZ German Research Centre for Geosciences

Keywords: Hydrogen storage; siliciclastic sediments; reservoir and sealing rocks; bio-geo interaction; laboratory experiments; numerical simulation; analogue study

---

1. Introduction

As a result of energy production from renewable sources like wind and solar power and to ensure a high electrical grid stability, fluctuations in energy supply have to be compensated by energy storage devices of different storage capacities. The largest natural storage potential is given by geological

---

* Corresponding author. Tel.: +49-3641-948608; fax: +49-3641-948622
E-mail address: henkel.steven@uni-jena.de.

Selection and peer-review under responsibility of the GFZ German Research Centre for Geosciences
doi:10.1016/j.egypro.2013.08.004

structures in the underground. In Germany, the storage of materials in geological formations is already applied for e.g. strategic reserves of natural gas, CO_2 sequestration and waste disposals. In the US and UK hydrogen is also stored in (salt) caverns [1] and can be also of importance for Germany in the framework of storing renewable energy, transformed in hydrogen.

However, due to their higher storage capacities and their widespread occurrence porous reservoirs (aquifers, depleted oil and gas reservoirs) might be even more relevant, especially in regarding long-term (seasonal) storage [2]. We present a brief introduction of the studied areas in the framework of the H2STORE project, which investigates the storage of hydrogen in siliciclastic formations and give an outlook of potential hydrogen reactions stored in sandstone reservoirs.

Nomenclature

XRF	X-ray fluorescence spectroscopy
ICP-OES	inductively coupled plasma optical emission spectroscopy
SEM	scanning electron microscope
XRD	X-ray diffraction
COPL	original intergranular porosity loss due to compaction
CEPL	original intergranular porosity loss due to cementation
qz	quartz
chl	chlorite
bit	bitumen
fsp	feldspar
ab	albite
ill	illite
cc	calcite
an	anhydrite
kaol	kaolinite

2. Study area and Methods

The aim of our study is to investigate the main clastic sediment storage formations and their sealing mudstones in Germany. Therefore samples of five different locations (Fig. 1) with different geological settings are investigated (Tab. 1). Numerous analytical methods (e.g. light microscopy, XRF, ICP-OES, SEM, XRD) are applied to study the chemical and mineralogical composition of the samples, with a special emphasis on the pore-exposed grain surfaces, which may have a great reaction potential with long term stored hydrogen.

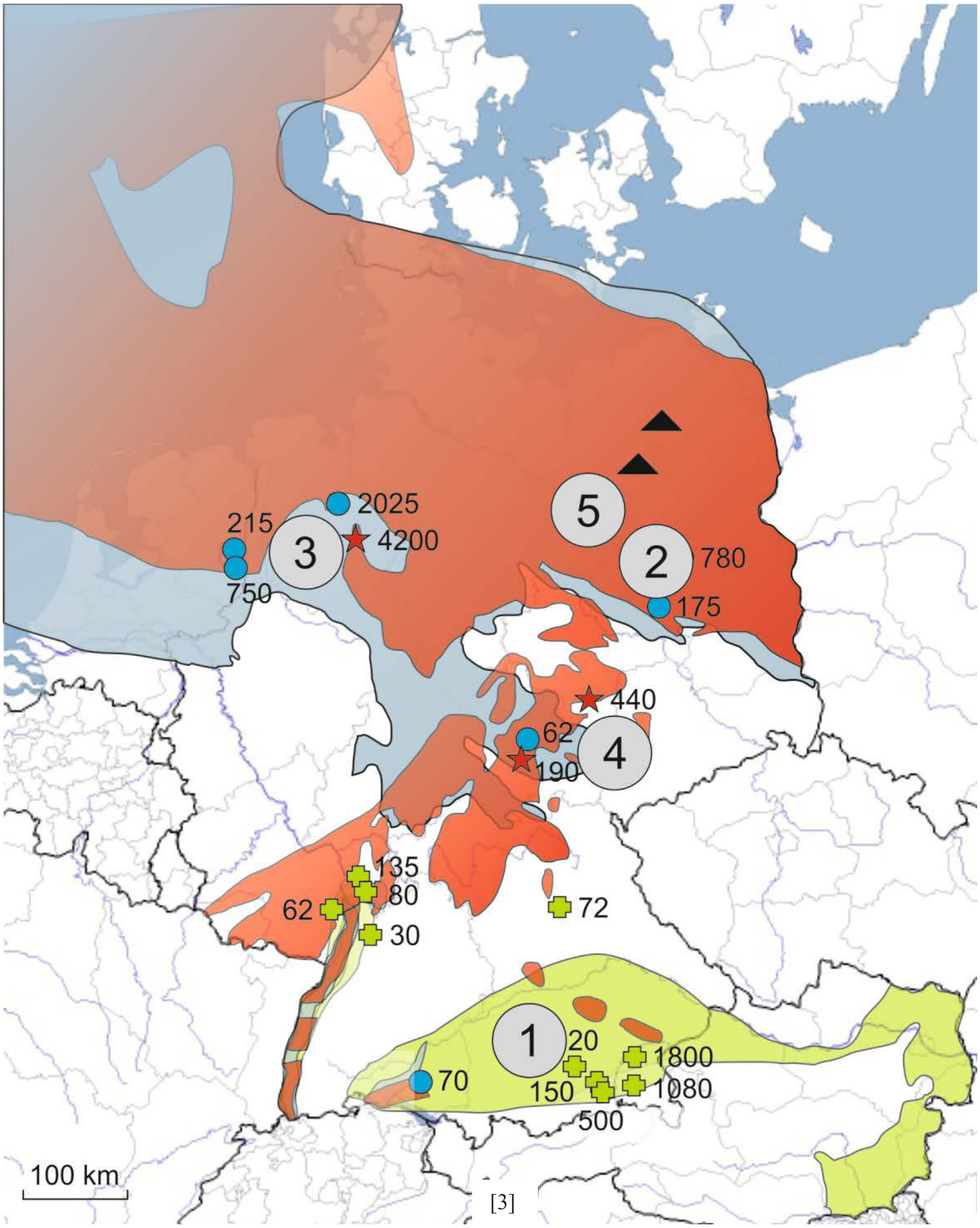

Fig. 1. Storage reservoir and research sites (No. 1-5, cf. Tab. 1) of the H2STORE project in Germany. In red = Permo-Carboniferous, in light blue = Early Triassic and in green = Tertiary strata. Symbols mark storage reservoirs in use (red stars, blue dots, green crosses) and under construction (black triangles). Numbers refer to storage capacities in Mio. m³ (modified after [4]).

Table 1. Overview of the H2STORE research areas (Fig. 1) and their geological/structural variations in reservoir setting.

	(1) Bavaria[5]	(2) Brandenburg[6]	(3) Lower Saxony[7]	(4) Thuringia	(5) Saxony-Anhalt[8]
Age	~ 25 Ma	~ 225 Ma	~ 250 Ma	~ 250 Ma	~ 270 Ma
Stratigraphy	Tertiary	Keuper	Early-Middle Buntsandstein	Early-Middle Buntsandstein	Rotliegend
Current depths	~ 1.600 m	~ 650 m	~ 1.700 m	~ 800 m	~ 3.500 m
Current temperature	~ 53-60 °C	~ 40 °C	~ 55-122°C	~ 40-80 °C	~ 125 °C
Facies	molasses (turbidites & debris flows)	fluviatile, shallow marine	playa platform	playa platform, fluviatile	playa platform
Lithology	heterogeneous (sand- & mudstone, carbonate clasts)	heterogeneous (sand-, silt- & mudstone)	heterogeneous (ooid-, silt-, sand- & mudstone)	silt- & sandstone	silt- & sandstone
Reservoir type	gas storage site	CO_2 storage site	gas storage site	depleted gas reservoir	depleted gas reservoir
Overburden	shallow marine limestone & clastic sediments	massive mudstone & carbonate	marine shale, sulphate, halite	marine shale, sulphate, halite	salt & carbonate

3. Results

The burial history of the different sample locations varies, with maximum burial depths of about 4.0-8.0 km in Saxony-Anhalt [8] and 2.8-1.8 km in Thuringia [9], most probably established during Triassic-Cretaceous and Neogene times.

First (sub-) microscopic analysis confirm significant detrital, mineralogical and petrophysical differences in the studied rocks. Due to depositional conditions and diagenetic evolution the analyzed reservoir samples vary in grain size from medium-grained to fine-grained sand- and siltstone with point grain to suturated grain contacts, reflecting different stages of sandstone evolution during reservoir rock evolution. All the reservoir sediments are sealed by overburden mudstones with commonly low amounts of silty components.

Based on the detrital mineral content, the studied samples show a widespread variability, but also some regional clusters. According to the classification of McBride [10] sandstones from the Brandenburg site are lithic arkoses to feldspathic litharenites. In contrast Tertiary samples studied in Bavaria are of litharenitic composition. Samples taken from similar Buntsandstein stratigraphic horizons reveal slightly different compositions in Thuringia and Lower Saxony, ranging from arkoses to lithic subarkoses (Fig. 2).

The compaction state in figure 3, after Houseknecht [11] and Ehrenberg [12] indicates a broad spectrum of mechanical compaction, cementation and porosity proportions. Permian (Saxony-Anhalt) samples show a larger influence of early cementation in contrast to the more mechanical compacted samples from Thuringia (Early Triassic).

All together the sandstone composition varies from subarkoses to litharenites with a highly variable content of pore filling cements and porosity. These features confirm the suggestions already made by observations of grain-contacts which imply different rates of rock compaction and therefore on the potential of pore space evolution and its diagenetic mineral filling.

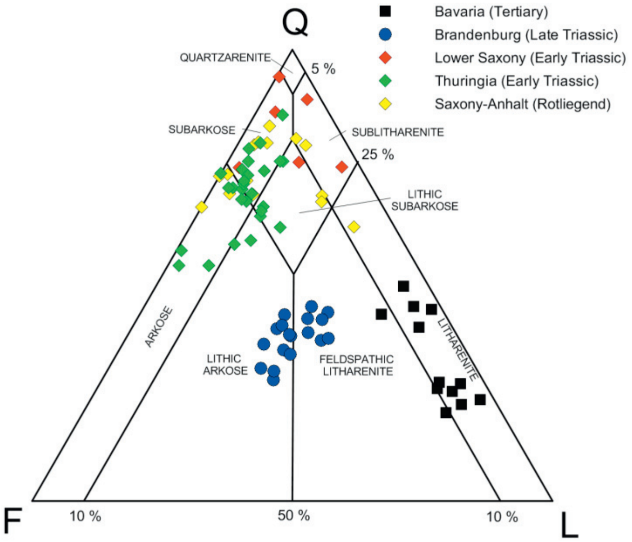

Fig 2. Variation in sandstone composition in terms of their quartz (Q), feldspar (F) and lithoclast (L) contents of the studied areas. Due to their different mineral contents the sandstones are classified as (sub-) arkoses to litharenites, following the scheme of McBride [10]. Data from ([5], [6], [7], [8], [13] and own unpubl. data).

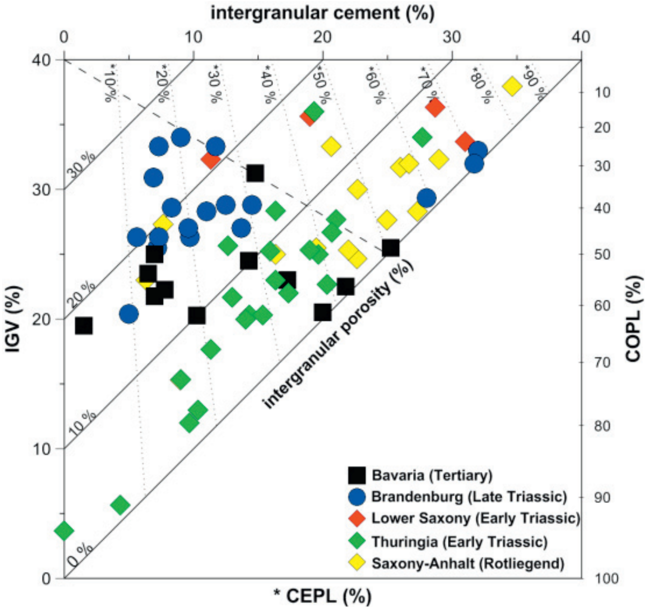

Fig 3. Pore space evolution in reservoir sandstones of the studied areas. Discrimination after Houseknecht [11] and Ehrenberg [12]. The Rotliegend samples show an early cementation on the pore space evolution due to their high amounts of intergranular cement with respect to the intergranular volume (IGV). The Tertiary and Late Triassic samples have a reduced pore space (low IGV values) along with low pore filling cement values, which indicates a higher mechanical compaction of the pore space. Note the Early Triassic samples: here the Lower Saxony samples have high cement content and only minor indications of pronounced compaction processes. In contrast the Thuringia samples have low cement content by higher rock compaction. These features suggest different diagenetic and tectonic evolution in these areas during post-Triassic time.

The major pore filling cements are carbonate minerals (Fig. 4J), anhydrite (Fig. 4H), quartz (Fig. 4H) and feldspar (Fig. 4G).

The presence of site- and facies specific authigenic clay minerals is shown in figure 4. Clay minerals that occur in the samples are identified as illite (Fig. 4E-F), chlorite (Fig. 4A-D) and kaolinite (Fig. 4I), coating detrital grains which are most frequenty exposed to the pore space.

Bitumen, as a remnant of hydrocarbons, is present in the depleted reservoirs (Fig. 4A) and most common in Permian samples. Chlorite is a typical component in the Permian of Saxony-Anhalt and ooids are numerous in the Buntsandstein of Lower-Saxony (Fig. 4J).

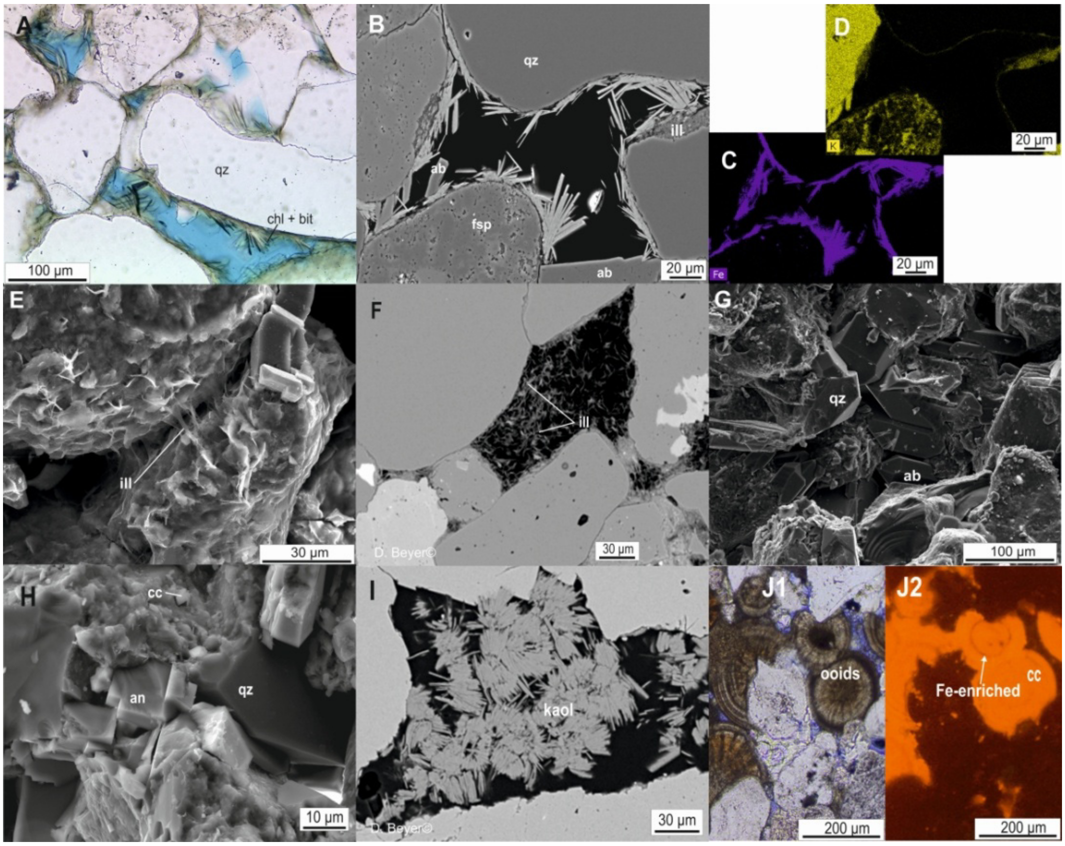

Fig. 4. Examples of the most common pore exposed mineral phases in the studied sites: in Saxony-Anhalt chlorite (Fig. 4A-D) and illite are abundant, whereas in Lower Saxony and Thuringia meshwork and fibrous illites are frequent (Fig. 4E-F). Additionally kaolinite (Fig. 4I) is present in some Thuringian sites. Pore-filling blocky cements, like carbonate, anhydrite, quartz, and feldspar (Fig. 4G, H) are widespread in all areas. However, in Bavaria carbonate is mainly formed during shallow marine deposition and forms ooids (Fig. 4J). A= light microscope; J= cathodoluminescence; all others SEM – Back scattered (B, F, I = thin section, E, G, H = rock fragments, C = element distribution by EDX (yellow = K , blue = Fe).

First petrophysical results are shown in figure 5 and vary between the different sample sites and sample locations. In general authigenic clay minerals present in the samples of the Saxony-Anhalt reduce permeability and porosity. Samples that contain large amounts of blocky cements (Lower Saxony) have minor porosity but slightly higher permeability levels.

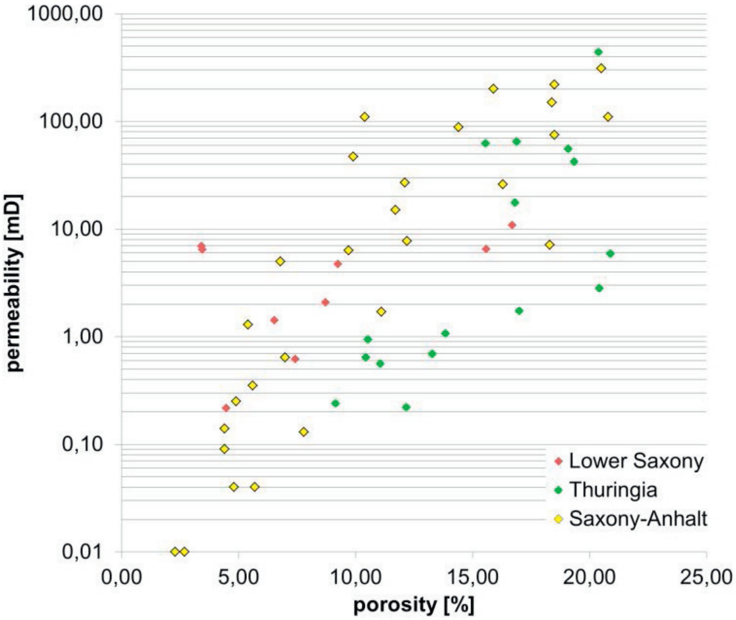

Fig 5. Porosity-permeability variations in the reservoir rocks of studied areas. In general all rock suites show an increase in porosity by enhanced permeability. Lower values are connected to higher contents of clay minerals and pore-filling cements and to mechanical compaction (cf. Fig. 3). These features reduce the pore space volume and prevent extensive fluid flow migration, resulting in moderate to poor reservoir conditions. (Data source: Saxony-Anhalt [8], Thuringia: D. Pudlo, unpubl. data, Lower Saxony: D. Albrecht, pers. comm.).

4. Discussion & Outlook

The detrital compositions of the rocks vary due to differences in source areas, transport conditions and depositional environments at the investigated sites. Additionaly post depositional, diagenetic burial evolution of the sandstones at different pressure, temperature and pH/eH/fluid conditions strongly affect fluid-rock interactions. Therefore the study on the mineralogical content of such different reservoir rocks will help to understand the influence of potential hydrogen reactions on reservoir quality.

Pore spaces, completely filled with blocky cements (e.g. anhydrite, quartz - Fig. 4H) exhibit reduced rock porosity. Due to this feature no or only reduced fluid/gas flow in these parts of the reservoir are expected.

The rock permeability is mainly related to clay minerals and their morphology, but these minerals may also strongly increase the pore-exposed reaction surface. This exposure will increase fluid-rock interactions. These features and the amount of blocky pore-filling cements (like e.g. carbonate, anhydrite) result in a general, mostly site-specific correlation of rock porosity and permeability. Thereby these fluid-rock reactions will be further complicated by the occurrence of e.g. hydrocarbons (bitumen, Fig. 4A) and iron compounds (Fig. 4C) which stain clast rimming clay minerals [14]. Moreover the composition of the formation waters in the depleted reservoirs (Fig. 6) and the presence of microbiological habitants [15] will influence any mineralogical/geochemical reaction.

Also the potential generation of bio-methane [16] and of “gas-fingering” processes [17] by the injection of hydrogen in the sandstone reservoirs have to be considered and requires further investigations [18].

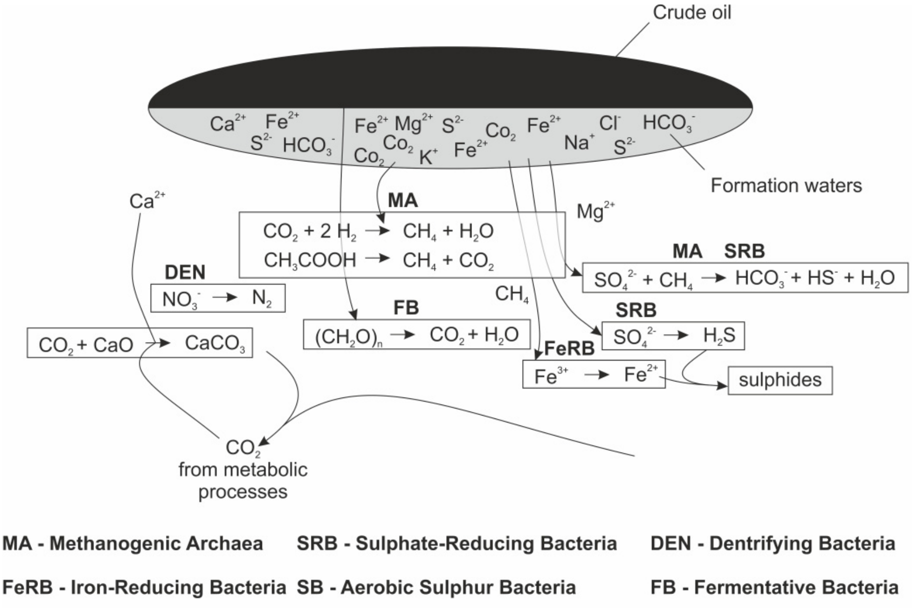

Fig. 6: Potential bio-(hydro-) chemical reaction scheme in hydrocarbon reservoirs [19].

Acknowledgements

H2STORE is part of the research and development programme “Energy Storage“. We thank the German Federal Ministry of Education and Research (BMBF) for funding (grant nos. 03SF0434A – FSU Jena, 03SF0434B – TU Clausthal, 03SF0434C – Helmholtz Centre Potsdam, German Research Centre for Geosciences) and E.on Gas Storage GmbH, GDF SUEZ E&P DEUTSCHLAND GMBH, Rohöl-Aufsuchungs Aktiengesellschaft (RAG)-Austria, and RWE Gasspeicher GmbH for their assistance.

References

[1] Stone, H.B.J., Veldhuis, I. und Richardson, R.N. Underground hydrogen storage in the UK. In: Evans, D.J. und Chadwick, R.A. (Hrsg.). Underground Gas Storage – Worldwide Experiences and Future Development in the UK and Europe. *The Geological Society, London, Special Publication 313*, 2009, p. 217-226.

[2] Lord, A.S. Overview of Geologic Storage of Natural Gas with an Emphasis on Assessing the Feasibility of Storing Hydrogen. *SANDIA Report SAND2009 – 5878*; 2009, p. 28.

[3] www.stepmap.de; 2012

[4] Reinhold, K., Müller, C., Riesenberg, C. Informationssystem Speichergesteine für den Standort Deutschland -Synthese-. *Bundesanstalt für Geowissenschaften und Rohstoffe*, Berlin /Hannover; 2011.

[5] Bottig, M. Diagenetische Umwandlung in Sandsteinen der gasgesättigten, der sekundär verwässerten, der Wasser-, und Übergangszone des Erdgas – Speichers Heidach in der Molasse Zone, Österreich. *Diploma-Thesis, University Vienna*; 2008.

[6] Förster, A., Schöner, R., Förster H.-J., Norden, B., Blaschke, A.-W., Luckert, J. et al. Reservoir characterization of a CO2 storage aquifer: The Upper Triassic Stuttgart Formation in the Northeast German Basin. *Marine and Petroleum Geology, 27*; 2010, p. 2156-2167.

[7] Nollet, S., Koerner, T., Kramm, U., Hilgers, C. Precipitation of fracture fillings and cements in the Buntsandstein (NW Germany). *Geofluids*, 9; 2009, p. 373–385.

[8] Pudlo, D., Reitenbach, V., Albrecht, D., Ganzer, L., Gernert, U., Wienand, J. et al. The impact of diagenetic fluid–rock reactions on Rotliegend sandstone composition and petrophysical properties (Altmark area, central Germany). *Environm. Earth Sci., 67*; 2012, p. 369-384.

[9] Götze, M. Numerische Modellierung der Versenkungsgeschichte der Thüringer Mulde mit Inkohlungsdates des Unteren Keuper. Diploma Thesis, Friedrich Schiller University Jena; 2011.

[10] McBride, E.F. A classification of common sandstones. *J. Sediment. Petrol 33;* 1963, p. 664-669.

[11] Houseknecht, D.W. Assessing the relativeimportance of compaction processes and cementation to reduction of porosity in sandstones. *AAPG Bull., 71;* 1987*, p.* 633-642.

[12] Ehrenberg, S.N. Assessing the relative importance of compaction processes and cementation to reduction of porosity in sandstones: Discussion; Compaction and porosity evolution of Pliocene sandstones, Ventura Basin, California:Discussion*. AAPG Bull., 73*; 1989, p. 1274-1276.

[13] Weibel, R., Friss, H. Opaque minerals as keys for distinguishing oxidising and reducing diagenetic conditions in the Lower Triassic Bunter Sandstone, North German Basin. *Sediment. Geol., 169*; 2004, p. 129–149.

[14] Foh, S., Novil, M., Rockar, E., Randolph, P. Underground hydrogen storage. Final report. *Brookhaven National Laboratory,* Upton, New York; 1979, p. 145.

[15] Morozova, D., Alawi, M., Shaheed, M., Krüger, M., Kock, D., Würdemann, H. The influence of microbial activity on rock fluid interaction: baseline characterization of deep microbial biosphere for Enhanced Gas Recovery in the Altmark natural gas reservoir. *Energy Procedia, 4*; 2011, p. 4633-4640.

[16] Panfilov, M. Underground Storage of Hydrogen: In Situ Self-Organisation and Methane Generation. *Transport in Porous Media, 85*; 2010, p. 841-865.

[17] Paterson, L. The implications of fingering in underground hydrogen storage. *Int. J. Hydrogen Energy, 8-1*; 1983, p. 53-59.

[18] Pudlo, D., Ganzer, L., Henkel, S., Kühn, M., Liebscher, A., Lucia, M. D., et al. The H_2STORE project: hydrogen underground storage – A feasible way in storing electrical power in geological media? Clean Energy Systems in the Subsurface: Production, Storage and Conversion. *Geomechanics and Geoengineering*; 2013, p 395-412.

[19] Wolicka A., Borkowski, A. Introduction for Enhanced Oil Recovery (EOR). *InTech.*; 2012, p. 113-142.