High-resolution 3D fabric and porosity model in a tight gas sandstone reservoir: A new approach to investigate microstructures from mm- to nm-scale combining argon beam cross-sectioning and SEM imaging

Guillaume Desbois a,*, Janos L. Urai a, Peter A. Kukla b, Jan Konstanty c, Claudia Baerle d

a Structural geology, Tectonics and Geomechanics, RWTH Aachen University, Locherstrasse 4–20, D-52056 Aachen, Germany
b Geological Institute, RWTH Aachen University, Wülferstr. 2, D-52056 Aachen, Germany
c Wintershall Holding AG, Friedrich-Ebert-Straße 160, 34119 Kassel, Germany
d Wintershall Holding AG, Erdölwerke Barnstorf, Rechterner Straße 2, 49406 Barnstorf, Germany

1. Introduction

Producing gas economically from unconventional sources is a great challenge. Tight gas reservoirs, with their low permeability and low porosity, have recently received much interest because of the very large reserves, which could be produced with suitable technology. Tight gas reservoirs are found throughout the world and occur in all large reserves, which makes observations and interpretation difficult due to limits in resolution and sample preparation. They are also valuable for future exploration (Holditch, 2006; Littke et al., 2008).

For improved recovery of these tight gas reservoirs, it is essential to quantify porosity at the pore scale. However, because pores in tight gas reservoirs are usually below 1 μm, it is difficult because of technical and preparation limits. Thus, the fundamental information about pore morphologies in tight gas reservoirs is missing, although pore morphology and connectivity is the key to correlate porosity and permeability data.

Traditional techniques for characterization of different aspects of porosity in sandstone are X-ray computer tomography (micro-CT) scanning (Honarpour et al., 2003; Tono and Ingrain, 2008), magnetic resonance imaging (MRI) (Pape et al., 2005), particle size analysis, point counting based on petrographic thin sections, environmental scanning microscopy (ESEM) (Tiab and Donaldson, 1996), X-ray diffraction (XRD) (Ward et al., 2005), X-ray fluorescence (XRF) (McCann, 1998), confocal scanning laser microscopy (CSLM) (Menendez et al., 2001) and mercury porosimetry (Favvas et al., 2009). The combination of these complementary methods is considered to give a robust characterization of reservoir properties of sandstones (e.g. Baraka-Lokmane et al. 2009) and in sandstones this has produced a growing body of literature of 3D pore models (with a resolution of a few micrometers) and based on these, numerical models of fluid flow through porosity, which accurately predict bulk properties such as Darcy Permeability (Sholokhova et al., 2009; Tölke et al., 2010).

There is a considerable body of literature on the evolution of porosity by compaction and diagenetic processes (Gaupp et al., 1993; Schöner et al., 2008; Zwingmann, 2008) and it appears that to investigate porosity at nm scale in tight gas reservoirs, SEM imaging is the most direct approach (Nadeau and Hurst, 1991; Ziegler, 2006). This method, however, is limited by the poor quality of the investigated surfaces (mainly broken or mechanically polished surfaces including the decoration of porosity by colored resin embedment), which make observations and interpretation difficult (Lanson et al., 2002; Schöner and Gaupp, 2005).

The recent development of Argon ion source milling tools which produce polished cross-sections of exceptional high quality offers a new alternative for high resolution SEM imaging of porosity at nanoscale (Desbois et al., 2009; Loucks et al., 2009; Holzer and Cantoni, 2010).
Until now, the Ar-BIB preparation technique was mainly used for metallic materials but not for polyphase geomaterials. On the one hand, because it does not induce mechanical polishing, the Ar-BIB technique is an alternative to the resin embedment technique for preparation of delicate samples, which are difficult to polish because of phases with different hardness. In addition, the large area which can be polished (up to 2 mm$^2$) in comparison to the micro-porosity investigated) by the Ar-BIB allows investigating numerous features in one cross-section and also allows quantitative stereological analysis (Underwood, 1970; Russ and Dehoff, 2000), which is very difficult when broken samples surfaces are investigated. Ar-BIB cross sectioning does not require water or epoxy impregnation, which could damage the clay minerals by re-wetting. Thus, the Ar-beam polishing technique provides rapid, damage-free, reproducible surfaces in comparison to the conventional preparation techniques (Desbois et al., 2009; Loucks et al., 2009).

Focused Ion Beam (FIB) cutting was also demonstrated to be able to prepare high quality polished cross-sections in clay-rich geomaterials (Holzer et al., 2007, 2010; Desbois et al., 2009; Dvorkin, 2009). However, this technique induces potentially more damage of the surface since it uses mainly Gallium source (Erdmann et al., 2006) and typical areas produced by FIB (about μm$^2$) are much smaller than those prepared by BIB (about mm$^2$). Nevertheless, because FIB guns provide “focused” beam and are now fully implemented in commercial SEM machines, FIB instruments allows precise targeting of the region of interest and productive serial-cross-sectioning (Holzer et al.; 2010), respectively.

BIB and FIB are complementary approaches to prepare high quality surfaces suitable for high resolution SEM imaging (Holzer and Cantoni, 2011).

This contribution describes the use of the BIB high-resolution technique for a better understanding of pore space geometries in 2D and, for the first time, in 3D on tight gas reservoir sandstone samples from the Rotliegend in the north-west Germany.

**2. Methods**

Four samples were prepared by slowly cutting with a miniature diamond saw using air as cooling medium, from drill-cores slowly dried during storage in air; and faces ground dry using carbide paper from 800 down to 1200 grade. A flat surface of these samples of about 1 cm$^2$ was cross-sectioned perpendicular to the bedding, using a stand-alone argon beam machine (cross-section polisher JEOL SM-09010, Erdmann et al., 2006) to produce high quality polished cross-sections of about 2 mm$^2$, removing a 100 μm thick layer which was possibly damaged. We used 6 kV acceleration, achieving currents of about 150–200 nA for 8 h of milling. The principle of this technique is resumed in Fig. 1. The polished cross-sections were then coated with carbon, suitable for SEM imaging and EDX chemical analysis. The SEM microscopes used are a JEOL JSM-7000 F with an EDX system (EDAX-Pegasus work station) and a Zeiss Supra 55 equipped also with an EDX detector (EDAX, Apollo 10). Typically, SEM (in SE and BSE modes) imaging and EDX studies were performed at 15 kV at a working distance of 10 mm. Porosity and fabrics were statistically evaluated using the PolyLX Matlab toolbox (Lexa et al., 2005; http://petrol.natur.

**Fig. 1.** The principle of BIB cross-sectioning. (a) the ion beam irradiates the edge of sample un-masked by the shielding plate to create high quality polished cross-sections suitable for SEM imaging. (b) Overview of a typical cross section performed by BIB cross-sectioning.

**Fig. 2.** BIB serial cross-sectioning experiment. (a) Overview of the original mechanically pre-polished sample imaged in BSE mode; the dashed white line indicates the region of interest selected for the serial cross-sectioning experiment. (b) Serie of BSE micrographs from top view after the five successive cross-sections. These micrographs are used to measure the thickness of each cross-section: the 2nd, 3rd, 4th and 5th slices have a thickness of 37 μm, 55 μm, 51 μm and 51 μm respectively. In total, the selected region of interest was serial cross-sectioned over a depth of 210 μm.
For one of the samples, the stand-alone argon beam machine was also used by following a serial cross-sectioning procedure in order to estimate the evolution of microstructures in 3D. For this particular experiment, we produced five successive slices of about 50 μm thick (Figs. 2 and 3). After each cut, each surface was imaged at low magnification (as a mosaic made of more than 40 single images taken at × 500 magnification) both using SE2 and BSE detectors to produce general overview of fabrics and phase density maps over the entire BIB cross-section. An additional high magnification picture (as a mosaic made of 600–1000 single images taken at × 3000 magnification) was also produced with an SE2 detector to map the porosity at pore scale within the entire BIB cross-section. This procedure was repeated for all of the five successive cross-sections. Single images were assembled using Autopano software (Giga 2; Kolor) while successive pictures of cross-sections were aligned and rectified using ArcMap 9.3 software (ESRI) enabling reconstruction of fabric and porosity in 3D. For this serial cross-sectioning experiment, the conditions of imaging are similar to those used for single BIB cross-sectioning experiments.

In order to compare our approach with conventional techniques of preparation (Fig. 4), we also prepared mechanically polished thin-sections suitable for optical transmitted light microscopy and electron microscopy (gold coated) as well as broken samples for electron microscopy (gold coated).

### 3. Geological setting of the samples analyzed

The studied core samples originate from a well drilled in the Arsten Graben, south of Bremen. The area is situated at the southern margin of the North German Basin, which forms part of the Southern Permian Basin (SPB) (Glennie, 1986, Glennie, 1990, George and Berry, 1993, Plein, 1995, Strömbäck and Howell, 2002, Legler, 2005). Both are integral part of the Central European Basin System (Littke et al., 2008).

---

**Fig. 3.** SE and BSE micrograph mappings of the five successive cross-sections produced for BIB serial cross-sectioning experiment. At the scale-view of SE micrographs, the largest intergranular volumes filled with hairy illite (black arrows) and fractures, which fit roughly the sand-grain contours (white arrows), are clearly visible. BSE micrographs show the evolution of mineral phases over the investigated volume. Darker gray value indicates quartz grains while lighter gray values indicate mostly calcite, feldspars and calcite at the scale of micrographs. The black square from the SE cross-section-2nd cut is a missing picture in the image set.
The SPB stretches with a width of 300–600 km over ~1.700 km from the eastern United Kingdom to central Poland and the Czech Republic. The Basin has an asymmetric shape with its deepest part in the North (McCann, 1998). It is composed of several NW–SE-trending en echelon subbasins like the Silverpit/Dutch, the North German and the Polish Basin, which are separated by N–S and NW–SEE Variscan basement highs (Börmann et al., 2006). In the Upper Rotliegend I, tectonic quiescence with subsidence being mainly driven by compaction and thermal relaxation prevailed, following a phase of intense Lower Rotliegend bimodal magmatism (van Wees et al., 2000). This changed with the onset of the Upper Rotliegend II, which saw a new phase of extensional faulting and associated sedimentation (Bachmann and Hoffmann, 1997).

The investigated reservoir rocks are from the Upper Rotliegend II Bahnsen Member of the Hannover Formation. In this sequence, sediments of preliminary fluvio-aeolian origin, including barchanoid dune, wet to dry interdune, braided fluvial and alluvial fan sediments were deposited with thicknesses ranging between 180 m and 450 m. The analyzed sandstone sample from approximately 4800 m depth is of aeolian dune origin and has a white/gray color, a porosity of 8.7% and a permeability of 0.2 mD. Porosity was measured on core plugs using Mercury injection porosimetry. The measured porosity is then related to the effective (connected) porosity, of a dried sample under lab conditions. The porosity and permeability measurements reported reflect the average values for the studied stratigraphic member (no cut off has been applied). The studied samples were selected from part of the core, which are considered as representative in regards to porosity and permeability as well as mineralogy. Interesting locations were first selected based on log-data; then homogeneity was checked in 2D in large thin sections in the optical microscope and in volume using μCT imaging. It has been observed during testing of the well, that the more favorable reservoir quality is below the gas–water contact, while in the gas-bearing zone above, the porosity/permeability is drastically reduced by authigenic minerals. This led to uneconomic flow rates and the well was subsequently plugged and abandoned.

4. Results

4.1. BIB cross-sectioning

All single BIB cross-sections (Figs. 1 and 3), produced at similar conditions (6 kV for 8 h. of milling), have comparable polished-surface sizes of about 2 mm wide and 1 mm long resulting in area up to 2 mm². Each single BIB cross-sections have a pseudo Gaussian shape reflecting the distribution of ion density in the beam. It was slightly convoluted by the rocking motion of the sample during milling used to minimize the curtaining of milled surfaces, which occurs typically for heterogeneous materials. Investigations of microstructures at high magnifications (Figs. 5 to 11) give evidence for extremely well-polished surfaces down to the resolution of SEM (≥3 nm) without damage or artifacts which may disturb the imaging of sub-micrometric structures. Therefore, the high quality of surfaces allows investigating microstructures down to the resolution of SEM in true 2D cross-sections (Figs. 5 to 11). The high precision of BIB cutting is particularly well illustrated by Fig. 7,h, which shows that BIB is able to cut even the hairy illite particles of 50 nm in diameter and 1–2 μm long along the direction of length.

Serial cross-sectioning (Figs. 3 and 11) is a major improvement of the use of stand-alone BIB machines since it allows us to investigate microstructural evolutions in 3D. As far as we know, stand-alone BIB machines were never used for serial-cross-sectioning until now.

The process of serial sectioning does not decrease the quality of the BIB cutting: surfaces have similar and reproducible quality and no subsequent waste redeposition was detected at scales of our observations. During the serial cross-sectioning experiment the slice
thickness was targeted to be 50 μm. In practice, slice thicknesses are in the range of 51–57 μm (Fig. 2) resulting in an inaccuracy of 6% from the target point. Thickness of the slices is accurately measured by imaging the top of the cross-section (Fig. 2). The inter-slice thickness (here about 50 μm thick) is too thick to give the exact interpolation of microstructures along successive slices. Using our BIB cross-sectioner, the slice thickness can be reproducibly reduced down to 20 μm.

4.2. Grain fabrics and mineralogy

The samples show the typical features of tight, diagenetically altered aeolian sandstone from this part of the Central European Basin (Gaupp et al., 1993). Optical thin sections (Fig. 4.a) and ion beam polished cross-sections (Fig. 5.a) show that the diameter of the quartz grains is about 0.2 mm with sub-angular to round grains. Angular grains are rare and were only observed as “floating” fragments in pores cemented by clay matrix (Figs. 7.e and 9.c). Grain fabrics are dominated by sutured and concavo/convex contacts possibly evolved by dissolution processes and grain re-arrangement during compaction. EDX mappings made both on mechanically-polished thin-sections and BIB cross-sections reveal similar mineralogy (Fig. 6). Quartz grains dominate largely the mineralogical composition. Other minerals such as calcite, halite and feldspars are also present but in much less amount.

Optical transmitted light microscopy (Fig. 4.a), EDX measurements and electron microscopy performed on broken surfaces (Fig. 4.b and c) and BIB cross-sections corroborate the presence of diagenetic clay minerals between the sand grains (Bashari, 1998). In the vicinity of non-clay minerals, these are predominantly illite while kaolinite is mainly present in the region surrounding the feldspars probably due to the replacement of original feldspar grains. Clay cementations are not post-dated by any other cement. In optical thin sections (Fig. 4.a), only the biggest primary pores at triple junctions (>10 μm in size), decorated in blue by resin impregnation, are identified. However, SEM observations on broken surfaces (Fig. 4.b) and mechanically polished thin-sections (Fig. 4.c) show that these primary pores are not simply voids but complex pore microstructures based on the phyllo-mineral arrangement filling the inter-granular pore space. Fig. 4.b shows that the illite displays two different morphologies: fibrous/hairy or platy. The poor quality of mechanically prepared surfaces (Fig. 4) does not allow the detailed characterization of clay-pore microstructures. In contrast, BIB-prepared cross-sections show details of the morphology of pores in clay down to 10 nm in size (Figs. 5 to 11).
Fig. 6. EDX mapping for chemical composition analysis. Grains forming the matrix are mainly quartz (Si + O) but calcite (Ca + C), feldspar (Si + K + Al) and halite (Na + Cl) are also significantly present. Large amount of clay materials, as illite (K + Al + Fe + Mg) and kaolinite (Al + Si), cements the sandstone. Yellow color gives the highest relative specie content and black the lowest. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Fig. 7. SE and BSE micrographs of pore microstructures at pore scale. (a) Pore located at junction formed by four detrital grains (three quartz grains in deep black and one feldspar grain in gray). This pore is early cemented by halite (white color) and postdated by illite cementation arranged tangential at the direct vicinity of the detrital grains edge and as “hairy/fibrous” toward the pore center. (b) Same pore presented in (a) showing the apparent surface porosity. (c) and (d) detail from (b). In hairy/fibrous illite, three types of porosity are visible in (d) (see the text for details). (e) Pore located at junction formed by three detrital quartz grains. This pore is locally early cemented by halite (white color) and postdated by illite cementation arranged tangential at the direct vicinity of the sand-grains edge and as “hairy/fibrous” toward the pore center. We see also euhedral quartz grain (Qtz*) overgrown from sand-grain and angular quartz grain as “floating” fragment (Qtz+). (f) Same region presented in (e) showing the apparent surface porosity. (g) Tangential illite coating a calcite grain serving of substratum for hairy/fibrous illite. (h) Pore space as appearing in hairy/fibrous material showing in detail the meshwork formed by illite fibers aggregates close to the center of the pore shown in (f). (i) Detail of a single grain–grain contact filled with illite: tangential is coating edges of the two adjacent sand-grains while the median part of the contact is filled with hairy/fibrous illite. A F1 fracture is passing through the illite filling at contact between tangential and hairy/fibrous illite. (j) Same region presented in (i) showing the apparent surface porosity. (k) Detail of apparent porosity as appearing in a large multiple grains junction. Edges of adjacent grains are coated by tangential illite while hairy/fibrous illite filled the center of the intergranular volume. Euhedral quartz overgrowth (Qtz*) are embedded into the illite-rich region.
and minor quartz species are involved in the early cementation of different mineralogical phases, it appears that calcite, halite, feldspar homogeneously distributed (Fig. 5.a, 7.a, 7.c, and 7.e) and calcite andfeldspars seem to fill simply the intergranular volume. Feldspar grains (and much less commonly calcite and quartz) exhibit clear evidence of diagenetic dissolution and alteration (Figs. 8 and 9). Hematite grains (see Fig. 8.a) are sometimes found in the clay matrix but rare; this is in good agreement with the white/gray color of the sample since hematite is responsible for the “red bed” color of Rotliegend sandstones (Torrent and Schwertmann, 1987).

4.3. Pore microstructures

In the following sub-sections, observations are only based on BIB cross-sections, which allow accurate and detailed investigation of pore microstructures at pore scale.

4.3.1. Fracture porosity

As shown in Figs. 3, 5 and 11, both samples are significantly fractured. Two kinds of fractures are identified. Both are open and never cemented. The first set of fractures (F₁) occurs at the boundaries of clastic grains, in the clay cement (Figs. 5.b and c) or cutting the clay–clastic grain contact. F₁-fractures have jagged-walls. Segmentation of the F₁-fracture network (on single BIB cross-sections) and analysis using Image J 1.38× (Abramoff et al., 2004) show that these fractures represent 1–2% of the total polished cross-sections. The second set of fractures (F₂) is located within the clastic grains, with straight walls and abutting against other fractures. F₂-fractures comprise 1–2% of the total polished areas. Thus, the total porosity by fracturing (F₁ + F₂) is estimated equal to 2–4% of the total surface revealed by BIB cross-sectioning.

It is important to note here that in the case of the true 2D sections studied here, the area fraction in the image is a good measure of the volume fraction (Underwood 1970; Russ and Dehoff, 2000). In sections with topography or finite thickness this relationship rapidly gets lost (Krabbendam and Urai, 2003).

Serial cross-sectioning experiment (Figs. 3 and 11) shows that fracturing affects the entire mass of samples and that F₁ fractures are particularly connected and promoted according to the distribution of illite cementation.

4.3.2. Non clay minerals

Minerals such as quartz, feldspars and calcite (Fig. 8) show evidence of intra-crystalline porosity. Feldspar commonly exhibits intra-crystalline porosity with the pores having a pseudo-cubic morphology, which appears to be controlled by the crystal lattice. They are homogeneously distributed and have a range size of 200–500 nm. In calcite and quartz pores are rare, and occur as small rounded holes of around 500 nm in diameter. Euhedral quartz grains occur as overgrowth on quartz grains (Figs. 7.e, 7.k and 11.c), halite fills locally the intergranular volume as “pervasive-intrusions” (Figs. 5.a, 7.a, 7.c, and 7.e) and calcite andfeldspars seem to fill simply the intergranular volume. Feldspar grains (and much less commonly calcite and quartz) exhibit clear evidence of diagenetic dissolution and alteration (Figs. 8 and 9). Hematite grains (see Fig. 8.a) are sometimes found in the clay matrix but rare; this is in good agreement with the white/gray color of the sample since hematite is responsible for the “red bed” color of Rotliegend sandstones (Torrent and Schwertmann, 1987).

4.3.3. Clay minerals

Fig. 7 presents images focused on illite-rich regions. SE images are used for topography investigations while BSE images give information about the nature of phases and details of phases’ arrangement. All prepared samples exhibit similar microstructures and pore morphologies in illite cementation, which occurs typically at grain contacts and filling the intergranular volume.

Non-clay minerals are systematically coated with illite minerals, which are always tangentially organized to the surface of the sand grain, with a compacted fabric and pores close to the limit of SEM resolution. When they are visible, these pores are about 500 nm long and less than 50 nm wide, elongated parallel to the adjacent surface of non-clay mineral.

By studying the relationships and microstructures between the different mineralogical phases, it appears that calcite, halite, feldspar and minor quartz species are involved in the early cementation of the sandstones. Euhedral quartz grains occur as overgrowth on quartz grains (Figs. 7.e, 7.k and 11.c), halite fills locally the intergranular volume as “pervasive-intrusions” (Figs. 5.a, 7.a, 7.c, and 7.e) and calcite and feldspars seem to fill simply the intergranular volume. Feldspar grains (and much less commonly calcite and quartz) exhibit clear evidence of diagenetic dissolution and alteration (Figs. 8 and 9). Hematite grains (see Fig. 8.a) are sometimes found in the clay matrix but rare; this is in good agreement with the white/gray color of the sample since hematite is responsible for the “red bed” color of Rotliegend sandstones (Torrent and Schwertmann, 1987).
Fig. 9. Distribution and segmentation of porosity from typical and representative porous regions. Tangential illite (I), hairy/fibrous illite (II), kaolinite (III), feldspar (IV) and bended tangential illite (V), different regions are outlined by red dashed lines. (a) Different adjacent porous regions (I+II+III+IV). The mineral at the down right corner is calcite. (b) Segmented porosity as appears in micrograph (a) used to quantify and describe statistically the porosity. Each region is encoded by a different color (see the legend on figure). (c) Typical evolution of porosity into a multiple grains junction region partially cemented by illite and floating angular quartz fragment. Illite becomes much more porous towards the center of pore until it developed a pronounced hairy fabric. (I + II + III + V) regions are distinguished. (d) Segmented porosity as appears in micrograph (c) used to quantify and describe statistically the porosity. Each typical region is encoded by a different color (see the legend directly on the figure).
Toward the center of illite-rich regions, illite becomes significantly more porous. These clay particles have hairy/fibrous morphology forming a muddle meshwork with a slight tendency to be elongated perpendicular to the tangential illite. Here, we distinguished 3 main types of pore morphology: (1) Type I — elongated pores between similarly oriented clay sheets, (2) Type II — crescent-shaped pores in saddle reefs of folded sheet of clay and (3) Type III — large jagged pores where the density of hairy/fibrous illite is less. Type III pores are typically >1 µm, Type II between 1 µm and 150 nm and Type I <150 nm.

In our samples, illite-rich regions are mainly distributed along sand–sand contacts as an interconnected network (Figs. 5.a, 6 and 10). The study of the serial cross-sectioning experiment (Figs. 3 and 11) shows the spatial variability of illite cement distribution in 3D, corroborating that illite cement is matching the outer part of grains forming the sand by filling all the intergranular volume. In detail, primary multiple grain junctions are mostly filled by hairy illite while single primary grain–grain contacts are typically fully filled by tangential illite.

Another type of clay was identified to be kaolinite, which is locally found in the region surrounding the weathered feldspar grains. Kaolinite-rich regions have porosity characteristics within the range of tangential illite and hairy/fibrous illite. Pore morphologies of kaolinite-rich regions are in the range of the Type I and II as described above for hairy/fibrous illite. However, here the typical pore sizes are significantly smaller: Type I between 400 nm–100 nm and Type I <50 nm. Fig. 9.a and b shows an example of the coexistence of illite and kaolinite clays.

4.4. Quantification of porosity in significant porosity regions

Based on observations of more than 50 positions in the sample, all the different types of pores related to identified specific minerals and regions described in Section 4.3 are very similar in all studied positions. Therefore, a detailed study of few selected positions (typical examples in Fig. 9) is taken to be representative for porosity content of the specific regions (Table 1). Further quantification of this is in progress.

Figs. 9.a and b show porosity in adjacent areas formed by the succession of tangential illite, hairy/fibrous illite, feldspar and kaolinite. The tangential illite has low porosity at the resolution of SEM (2%). Kaolinite has a typical porosity of 5% and pore size of 0.8 ±3 µm; feldspars have a porosity of 16% and a pore size of 1.3 µm. The hairy/fibrous illite is the most porous region (24% of porosity in regions exclusively filled with hairy/fibrous illite) and pores around 1.3 µm. Generally pores have elongated shape (axial ratio greater than 2) but less pronounced for feldspars and kaolinite. The shape of pores in illite is individually controlled by the orientation of clay sheets at contact forming pores of Type I, II or III as described in the Section 4.3.3 (Desbois et al., 2009).

Fig. 9.c and d show a second typical porous region for primary multiple grain junctions formed by three grains of quartz partially cemented by illite and a floating angular quartz fragment. Averaged total porosity in multiple grain junctions is about 33%. Here, illite is organized tangentially at edges developing more and more hairy towards the center of the triple junction. Between these two end-members, illite trends to become hairy by bending its tangential organization according to the curvature of the quartz edges. This gradual development of illite microstructures is observed and quantified by the systematic increasing of the mean pore size and the total porosity contribution towards the center (Table 1).

5. Discussion

Microstructural observations presented in this contribution are in good agreement with the previous studies about illite characterization in reservoir sandstones. New insight is gained of porosity in clay-rich diagenetically altered samples by the preparation of extreme high quality, true 2D surface, revealing unprecedented detail of pore space and allowing quantitative analysis of the microporosity. Moreover, the use of the BIB instrument to produce serial cross-sections allows characterizing the fabric in 3D.

5.1. Microstructures and diagenesis

Though the aim of this contribution is not the study of diagenetic processes occurring in Rotliegend tight gas reservoir sandstone, we recognized many of the twelve diagenetic types distinguished in Gaupp (1996) and Schönér (2006). Following the interpretations in Gaupp (1996) and Schönér (2006), studied samples give evidence for the following phases in chronological order: Sekhia type (SB), illite coating type (IC), Hematite type (H), Feldspar leaching type (FL), Kaolinite type (K), Illite meshwork type (IM) and possibly the late quartz type (Q) for only the Bahnse sample. Thus, our observations are in good agreement with the general model of Rotliegend sandstone diagenesis (Macchi, 1987; McCann, 1998; Lanson et al., 2002; Ziegler, 2006; Abraham, 2007), and are interpreted to represent common rock and diagenesis types from the Rotliegend.

5.2. Fracturing

The morphology of F1-fractures indicates that these fractures are late and were formed by relaxation of stress (from over 200 MPa to atmospheric) after drilling by core damage (Holt et al., 1994).

Although intra-crystalline fracturing in sandstones due to compaction is a common process (Chester et al., 2004), the F2-fractures are not compatible with the burial history because they are free of cementation or pressure solution. This indicates that F2-fractures also formed late by core damage (Holt et al., 1994; Haimson, 2007).

Core damage is a common problem when evaluating reservoir porosity and correction for core damage is an as yet unsolved question. Using the technique presented here, the porosity due to core damage can be quantified by image analysis.

5.3. Intragranular volume filled by Illite

In sedimentary basins, illite is present as detrital matrix and as diagenetic cement. Diagenetic illite forms after burial to significant
depth (Macchi, 1987; Meunier and Velde, 2004; Schöner et al., 2008). Fibrous illite growth in the Central European Basin sandstones is episodic as determined by K–Ar measurement (Wilkinson and Hasseldine, 2002), rapid as suggested by their elongated morphology (Mullin, 1961) and their nucleation is almost exclusively upon preexisting illite grain coating (Pollastro, 1985; Whitney and Velde, 1993). Illite growth is a major factor in reducing the porosity and permeability of reservoir rocks (Stadler, 1973; Seeman, 1979; Kantorowicz, 1990) and cause major problems during enhanced recovery (Kantorowicz et al., 1986). Until now, direct quantitative analysis of the morphology of illite cement and its effect on permeability (Palat et al., 1984; Ziegler, 2006) has been difficult, because it was not possible to prepare 2D cross sections and 3D models of porosity (Underwood, 1970; Desbois et al., 2009) and because it was not clear to what extent the fabric of the illite is altered by the methods used to clean and prepare the samples for analysis (Rahman et al., 1995), or by the analysis itself (Hildenbrand and Urai, 2003).

After drilling, the core samples studied by us were only subject to slow drying in air. Critical point drying (Huggett, 1982; De Waal et al., 1986; Nadeau, 1998) to eliminate the capillary forces during drying which are known to possibly change the fabric of the thin illite fibers, was not possible.

In the studied samples, illite is the major clay mineral cementing the grain fabric and filling the intergranular volume (also primary porosity). Illite is defined either by fibrous or platy morphology (Seeman, 1979; Huggett, 1982; Nadeau et al., 1985). Illite cementation reduces the intragranular volume by more than 60% in multiple grain junctions and by more than 95% at single grain–grain contacts. These differences are attributed by different illite particle organization: (1) in multiple grains junctions, adjacent grains are coated by very compact and poorly porous tangential illite (about 5 μm thick) while much more porous and connected hairy/fibrous illite fills the remnant space of the intergranular volume contributing to 70% of porosity in this particular regions; (2) at single grain–grain contacts, intergranular volume is exclusively filled by poorly porous tangential illite.

From a 3D point of view given by the serial cross-sectioning experiment (Fig. 11), intragranular volume appears then fully filled by illite. When the intragranular volume is located at multiple grain junctions, hairy illite is able to develop toward the center of the primary pore from tangential illite substrate and results in relative highly porous regions. When intragranular volume is located at only two grain interfaces, hairy illite is rare and the intragranular volume is then entirely filled by tangential illite and results in very low porous regions. Therefore, the intragranular volume results in a network of large intragranular volumes mainly filled with relative high porous hairy illite connected by poorly porous “intragranular volume throats” filled with tangential illite.

According to Bushell (1986), illite arranged tangential to the grain surfaces has less effect on permeability than perpendicular clay minerals and hairy/fibrous illite has a lesser effect in reducing porosity than tangential illite.

5.4. Estimation of total porosity from micro-investigations — geometric homogenization

Because our approach combining Ar-beam cross-sectioning and high-resolution SEM imaging enables the detection of pore microstructure in a high quality true 2D cross-section, porosity at pore scale can be segmented reliably and then quantified and statistically interpreted. This has the potential to model porosity by a combination of characteristics at the scale of sand grains (intergranular volume) with information at much finer scale to predict with macro-scale pore-permeability properties.

Microstructures were linked to bulk porosity by using a simple porosity homogenization since porosity is only based on geometric considerations: the total porosity was estimated by the sum of individual porosity content of typical representative porous regions (examples in Fig. 9) convoluted by the percentage amount of each of these regions (Table 1).

Based on point counting performed on thin sections, our sample consists mainly of quartz (52%), calcite (10%), feldspars (6%), kaolinite (4%), illite cementation at narrow grain contacts (10%), big pores at triple junction (blue epoxy in Fig. 4.a) cemented by illite (15%) and fractures (average of 3%, see section 3.3.1). The porosity of these different typical representative porous regions was inferred from segmentation of porosity from our microstructural investigations at pore scale (Table 1). Quartz and Calcite are assumed non-porous and we neglected other minor regions known to occur in these samples. Table 1 summarizes data used for the estimation of the total porosity.

The total porosity (9.27%) estimated for the studied sample is in good agreement with the bulk porosity (8.72%) of a similar sample measured on a core plug of about 10 cm³. At first, our study shows that illite filling the intra granular volume and fractures both
contribute mainly to the porosity (table 1). In second, our microstructural investigations at pore scale show also that illite cements and fractures form an interconnected network. Therefore, this is proposed to explain why the total porosity estimated by our microstructural investigations (9.27%) matches very well with the effective (connected) porosity measured by mercury porosimetry (8.72%). The slight discrepancy between measured bulk porosity (8.72%, which is indeed the connected porosity) and our estimated porosity (9.27%, i.e. apparent porosity in 2D cross-sections) may be due to local heterogeneities in the sample which we missed in our microstructural observations, errors in pore counting, or/and due to porosity in feldspar grains (contribution to total porosity estimated around 1%, Table 1) since our estimation includes it implicitly as connected to clay minerals (Tangential and hairy/fibrous illite + kaolinite) which are assumed to be connected and contributing in majority to the overall permeability. Anyway, this discrepancy is lower than 5% and could be considered as acceptable. Because in our estimation quartz and calcite grains were assumed non-porous though few intra-crystalline pores were detected (Fig. 8) and because measured and estimated porosity are in the same range, these suggests that pores in quartz and calcite are poorly connected to the overall fabric and thus do not contribute significantly to the effective porosity.

5.5. Fabric and pore 3D-model

This contribution gives a first impression of the complex micro-porosity at pore scale in tight gas sandstones. Fig. 12 presents a 3D model of fabric and porosity occurring in the studied sample (Rotliegend sandstone, Bahnsen member, Arsten graben, Bremen, Germany) compiling all microstructural information detailed above. The model (Fig. 12) presents a Bahnsen sample with grain matrix comprising quartz, calcite and feldspar partially weathered into kaolinite. The intergranular volume, corresponding to initial free-space between the grains forming the matrix, is cemented by euhedral quartz overgrowth, halite and calcite; the remnant free space is fully filled by illite. Tangential illite coats systematically grains forming the matrix over a thickness of 1–5 μm; at multiple grains junctions, there is enough free-space left for hairy/fibrous illite development. Therefore in 3D, the intergranular volume appears then fully filled by illite with highly porous regions (corresponding to hairy illite at multiple grains junctions) connected by poorly porous “intergranular volume throats” (corresponding to tangential illite at single grain–grain contacts).

SEM micrographs were added in this model to reflect the diversity of typical pore morphologies observed in our samples. Feldspar grains commonly exhibit intra-crystalline pores with pseudo-cubic morphology. In calcite and quartz, pores originate from fluid inclusions appearing as small rounded holes. Some bigger pores occur also in calcite grains with more jagged morphology and filled with visible hairy/fibrous illite. Pores in tangential illite are elongated and laying between two clay sheet aggregates, while hairy/fibrous illite displays 3 types of pores whose shapes are controlled by the organization of clay sheet aggregates (Type I, II and III). Kaolinite-rich regions bear pores with similar morphologies as found in hairy/fibrous illite regions (Type I and II) but with smaller pore size.

Fractures identified in our investigations were not included in this model since they are not primary but induced by core sample collection. This 3D view of porosity fabric in a tight gas reservoir presents the concept of porous fill of the Intergranular Volume with relevant implications for fluid flow simulation in sandstone reservoir. The lattice-Boltzmann method was applied successfully on clean sandstone (Sholokhova et al., 2009) with intergranular volume free of clay cements. For tight gas reservoir sandstone, this simulation approach is more challenging since the intergranular volume is cemented with permeable clay (Tölke et al., 2010). The model presented here forms the basis for incorporating the transport properties of the pore fill into fluid flow simulations in tight gas reservoirs, by assigning representative permeabilities to the different diagenetic pore fills of the Intergranular Volume, and incorporating this information in network models of the intergranular volume as determined by micro-CT.

![Fig. 12. Fabric and porosity 3D-model in an idealized tight gas sandstone reservoir from Rotliegend formation (Bahnsen member, Arsten Graben, south of Bremen, Germany). This model compiles all microstructural information detailed in this contribution. See text for details.](image-url)
6. Conclusion

The use of the Argon beam for polished cross-sections preparation produces very high quality sections without damage and artifact to investigate quantitatively the pore network in tight gas sandstones at the state-of-the-art SEM resolution. This method has the potential to bring out a new concept for porosity investigation in tight gas sandstones by bridging the information based on microstructures at pore scale with macro-scale properties. It offers also the basis for a fundamental understanding of pore geometry, fluid flow and, in combination with cryogenic techniques (Holzer et al., 2007; Desbois et al., 2008; Desbois et al., 2009) will offer insight in the geometry and wetting characteristics of in-situ fluid phases in sandstone reservoirs.

Acknowledgments

This paper reports work done in the Wintershall Tight Gas Consortium at RWTH Aachen University. We thank Wintershall Holding AG for help with obtaining the Rotliegend sandstone core sample, and for funding the project. We are grateful to Alexander Schwedt (GFE at RWTH Aachen University, Aachen, Germany) for technical support for the SEM imaging, Werner Krauss and Christian Diebel (†) for preparation of the samples; Katharina Albert, Jochen Hürtgen and Prokop Zavada for image processing and Simon Virgo and Max Arndt for help with Autoptano and ArchMap softwares.

References


Diebel (†) will offer insight in the geometry and wetting characteristics of in-situ fluid phases in sandstone reservoirs.

References


Diebel (†) will offer insight in the geometry and wetting characteristics of in-situ fluid phases in sandstone reservoirs.