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# The use of X-ray computed microtomography for graptolite detection in rock based on core internal structure visualization

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ABSTRACT:

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This paper presents for the first time X-ray computed microtomography (µCT) analysis as a technique for Silurian graptolite detection in rocks. The samples come from the Jantar Bituminous Claystones Member of the Opalino core, Baltic Basin, northern Poland. Images were obtained with spatial resolution of 25 µm, which enabled the authors to create a 3-D visualization and to calculate the ratio of fissure and graptolite volume to the total sample volume. A set of µCT slices was used to create a 3-D reconstruction of graptolite geometry. These µCT slices were processed to obtain a clearly visible image and the volume ratio. A copper X-ray source filter was used during exposure to reduce radiograph artifacts. Visualization of graptolite tubaria (rhabdosomes) enabled *Demirastrites simulans* to be identified. Numerical models of graptolites reveal promising applications for paleontological research and thus for the recognition and characterization of reservoir rocks.

**Key words:** Computed microtomography; Visualization; Graptolites; Silurian; Baltic Basin shale.

## INTRODUCTION

Graptolites (Graptolithina) are extinct colonial marine animals that lived from the middle of the Cambrian to the uppermost Carboniferous. They were widely distributed in marine waters and their evolution was extremely rapid. Planktonic graptolites had the ability to vary the shapes of tubaria, thecae and genicular processes to fill a wide range of niches during the Ordovician and Silurian periods. Their complex morphology permits the establishment of many species in succession in sediments around the world. Thus graptolites are important index fossils and provide the primary means of correlation of Ordovician and Silurian strata (e.g., Štorch 2006; Zalasiewicz *et al.* 2009; Lenz *et al.* 2012).

Graptolites are studied from specimens either flattened on bedding planes or isolated after rock dissolution. The preservation of graptolites on the rock surface in many cases is not sufficiently good for taxonomic classification. The scanning electron microscope (SEM) is the most advanced tool for studying the detailed morphology of well-preserved specimens. However, dissolving rock in acid is not always possible or successful with fragile graptolites, as they may be partly or completely destroyed. X-ray investigation of graptolites in rock has so far only been used by Bjerreskov (1978) to identify possible soft body tissue.

In this paper we present the use of X-ray computed microtomography (μCT) as a non-destructive method for visualization of graptolites assemblages in successive parts of a well core plug, showing directly the original distribution of graptolites within the sediment. This method, when enhanced, could also prove useful for the purpose of detailed study of graptolites tubaria.

Black shales are typically associated with abundant graptolites, characterized by high TOC contents reaching Corg = 2–18%. Based on data from many studies, graptolitic shales are one of the main hydrocarbon sources that formed oil and gas fields in Paleozoic deposits around the world, e.g., Silurian graptolitic shales make up to 9% of all hydrocarbons that form the oil and gas fields in the largest petroleum basins (Klemme and Ulmishek 1991). The sedimentary organic matter in the shales is primary type II kerogen, which was partially derived from planktonic graptolites fauna (Berry 1998). Thus they are important as an indicator of the richness of organic matter in anoxic units – the sources of hydrocarbons in rocks. The main accumulation areas of graptolite shales were the margins of paleocontinents, e.g., Baltica (Finney and Berry 1997).

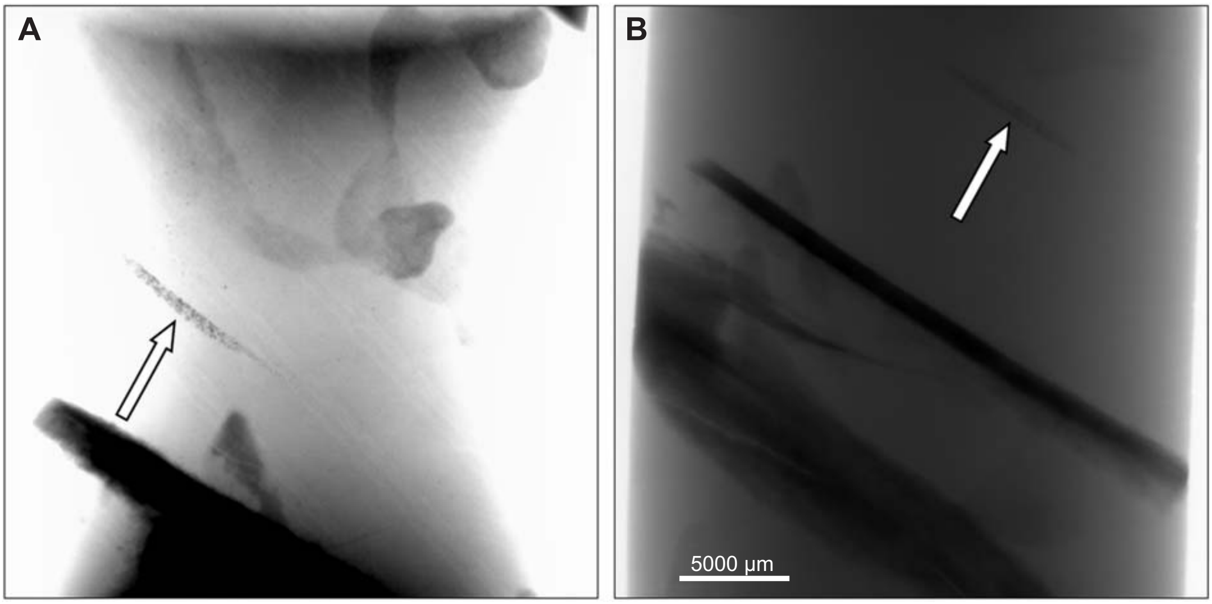
In the present study, a numerical model of Silurian graptolite fossils in black shale based on μCT images has been developed. Black shales have low transparency to X-ray radiation, thus a method was developed to obtain μCT images of the internal structure of the shale. The main purpose of this paper is to show the possibility of detecting the presence of graptolite assemblages in core samples using X-ray computed microtomography (μCT) and the possibility of their classification to the species level. This method can be useful for biostratigraphy as well as further paleontological studies, and so it could form an important component of sedimentation conditions analysis of hydrocarbon reservoir rocks.

## MATERIAL

The five samples selected are from the lowermost Silurian Jantar Bituminous Claystones Member in the Opalino core of Poland, located in the deeper neritic zone of the Baltic Basin on the western slope of the East European Craton. The samples are in the shape of cylinders with diameter of ~25 mm and height of ~60 mm, cut from a borehole core. Shale sedimentation took place in small subsidence zones. The Jantar shales have the highest organic carbon content (TOC) in the whole Silurian (Modliński *et al.* 2006; Podhalańska 2013, 2014). Graptolites were observed in two of the five samples (sample No. 1 and sample No. 2). The analyzed samples come from the following depths: 2871.40 m (sample No. 1), 2873.95 m (sample No. 2), 2875.75 m (sample No. 3), 2877.15 m (sample No. 4) and 2881.98 m (sample No. 5) below the terrain surface, and they represent the Aeronian, Llandovery. Detailed analysis of these five samples from the Jantar Member, Sasin Member and Piaśnica Member are presented in Kaczmarek *et al.* (2015) and Kaczmarek and Wejrzanowski (2016). According to this study the bulk densities of the samples (determined by buoyancy method) were in the range 2443 to 2580 kg/m$^3$. The elastic parameters were determined using non-destructive ultrasonic tests. The dynamic Young's modulus values were in the range 21 to 57 GPa and the Poisson ratio values 0.21 to 0.40 (Kaczmarek *et al.* 2015).

## X-RAY COMPUTED MICROTOMOGRAPHY (MCT) METHOD

The microtomography method is based on acquiring a series of X-ray images of the sample. The material is rotated at a constant angle and a radiograph is taken in each position. All scanned radiographs can be converted after the reconstruction process into one spatial numerical model, which represents the internal structure of the analyzed sample. This process is possible due to differences in the value of the linear absorption coefficient of the studied material. The linear absorption coefficient depends on the atomic number of the elements building the sample. The theoretical basis of μCT is described by Ketcham and Carlson (2001), Baker *et al.* (2012) and Cnudde and Boone (2013). An example of the standard use of μCT in the study of the microstructure of porous reservoir rock has been presented by Appoloni *et al.* (2007). Furthermore, the relation between μCT image resolution and the quantification process (data acquisition, radiograph reconstruction and image processing) of geometric parameters has been shown in Machado *et al.* (2014). A new tool for μCT research is the synchrotron. Bielecki *et al.* (2013) described the use of synchrotron-radiation-based computed microtomography together with laboratory-source-based microtomographic facility for data acquisition. The results can be used to determine physical properties (e.g., porosity, specific surface area) and diffusion tortuosity. Moreover, Fusseis *et al.* (2014) provided detailed guidance concerning the tomography method based on synchrotron together with a step-by-step description.



Text-fig. 1. Example of radiographs without (A) and with (B) copper filter of the sample from Milowo core, depth 3784.50 m, Jantar Bituminous Claystones Member. Milowo is located 30 km south of Opalino. White arrows show the same graptolite from separate µCT tests

In this study, the claystone samples of the analyzed Jantar bituminous shale have a high absorption factor, causing an unsatisfactory initial scanning of µCT. In further studies a 1.5 mm thin copper filter for the X-ray source was used to reduce the artifacts (beam hardening). This improved the quality of the images (Text-fig. 1).

The acquisition, which took 2 hours 51 minutes, generated data matrices of 1024 × 1024 pixels. Afterwards a total of 1024 µCT slices were obtained from the sample. Images were used to create a representative 3-D volume of the black shale.

Shale samples were examined by Xradia MicroXCT-ray with a Hamamatsu L8121-03 source, which generates X-rays in the range of 40 to 150 kV. A CCD video system was used to convert the images obtained to digital data with a resolution of 1024 × 1024 pixels and a 16 bit image depth of detection. A large field of view (LFOV) lens with 0.5 zoom was used during tests to obtain the broadest area of recognition. The parameters used for examination of samples with graptolites are shown in Text-fig. 2.

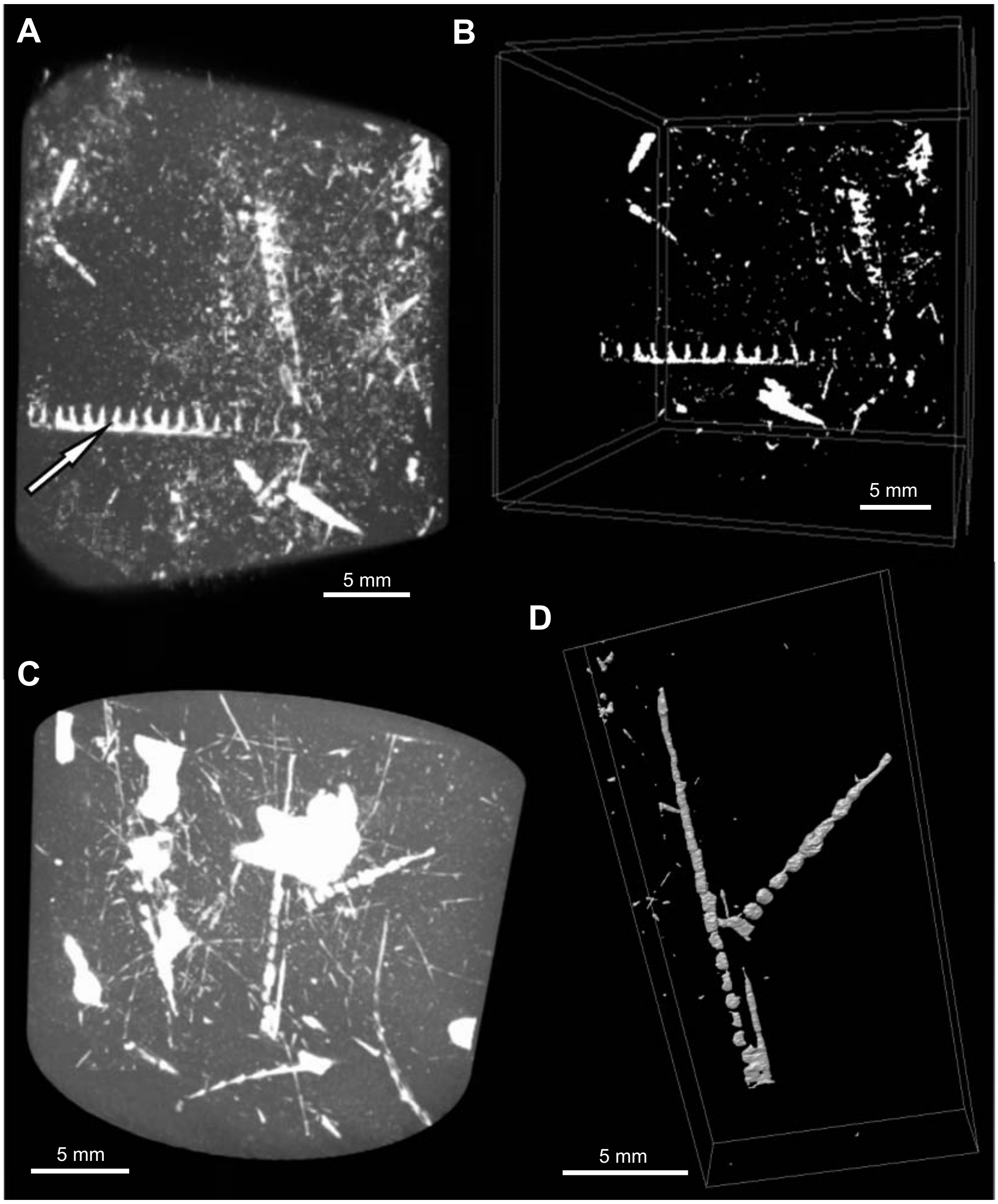
Due to the low X-ray radiation transparency of shale, the selected samples were scanned several times to obtain accurate images. After reconstruction, the images were processed from 16 bit to 6 bit to reduce the size of images for faster processing and then the thresholding method was used to binarize all the images. Graptolites and fissures were among the characteristic elements identified from the processed images. The selected working area, called ROI (region of interest), was prepared and smoothed by using 3D median and despeckle filters. In addition, during image processing, the ratio of graptolite and fissure volume to the total volume of the rock sample (Vv) was calculated.

## GRAPTOLITES

Several fragments of graptolites were detected, mostly straight monograptids, from depths 2871.40 m and 2877.15 m of the Opalino core. The 3-D µCT images of the graptolites are presented on Text-fig. 3.

| Sample | Voltage [kV] | Intensity [µA] | Exposure time [s] | Type of objective | Type of filter | Pixel size [µm] |
|---|---|---|---|---|---|---|
| 1 | 140 | 90 | 5 | LFOV | 1.5 mm Cu | 25 |
| 2 | | | | | | |

Text-fig. 2. Specification of X-ray microtomography acquisition and reconstruction



Text-fig. 3. 3-D µCT images and models of sample 1 depth 2871.40 m (A, B) and sample 2 (C, D) depth 2877.15 m, Opalino core, with graptolites. A – 3-D µCT image showing internal structure. B – 3-D model with *Demirastrites simulans* (Persen) arrowed, voxel resolution 25.5 µm. C – 3-D µCT image revealing the structure of sample 2, with densely packed graptolites, mostly thin forms. D – 3-D model with cross section of two monograptids, voxel resolution 25.3 µm

Classification of graptolites to genus or species level is possible when the characteristic morphological features are well preserved. One of the graptolite fragments from sample 1 differs, as it possesses isolated, characteristic metathecae arranged in the mode typical for *Demirastrites* (Text-fig. 2A, B). The measurements of this specimen indicate the distal part of the tubarium of *Demirastrites simulans* (Persen) with 14 thecae. The genus *Demirastrites* is characteristic of the Aeronian, Llandovery (Tomczyk 1974; Štorch and Kraft 2009; Radzevičius 2013). *Demirastrites simulans* (Persen) occurs in the *convolutus* bio-

| Series | Stages | Biozones |
|---|---|---|
| LLANDOVERY | Telychian | *Cyrtograptus insectus* |
| | | *Cyrtograptus lapworthi* |
| | | *Octavites spiralis* |
| | | *Monoclimacis crenulata* |
| | | *Monograptus crispus* |
| | | *Spirograptus turriculatus* |
| | Aeronian | *Spirograptus guerichi* |
| | | *Stimulograptus sedgwickii* |
| | | ***Lituigraptus convolutus*** |
| | | *Pri. leptotheca–M. argenteus* |
| | | *Demirastrites pectinatus–triangulatus* |
| | Rhuddanian | *Coronograptus cyphus* |
| | | *Orthograptus vesiculosus* |
| | | *Parakidograptus acuminatus* |
| | | *Akidograptus ascensus* |

Text-fig. 4. Llandovery, Silurian graptolite zonal schemes (after Melchin *et al.* 2012) showing the position of the *convolutus* biozone in the Aeronian. Abbreviations: *Pri. – Pristiograptus, M. – Monograptus*

zone, e.g., in the Kolka core in Latvia, Baltic Basin (Loydell *et al.* 2010) and *simulans* and *convolutus* biozones in Bohemia (Štorch 1994). Thus the depth of 2871.40 m of the Opalino core, with *Demirastrites simulans* possibly corresponds to the *convolutus* Biozone (Text-fig. 4).

## RESULTS

Analysis of the µCT images yields a 3-D visualization depicting graptolites from Silurian black shale from the Opalino core, Polish part of the Baltic Basin.

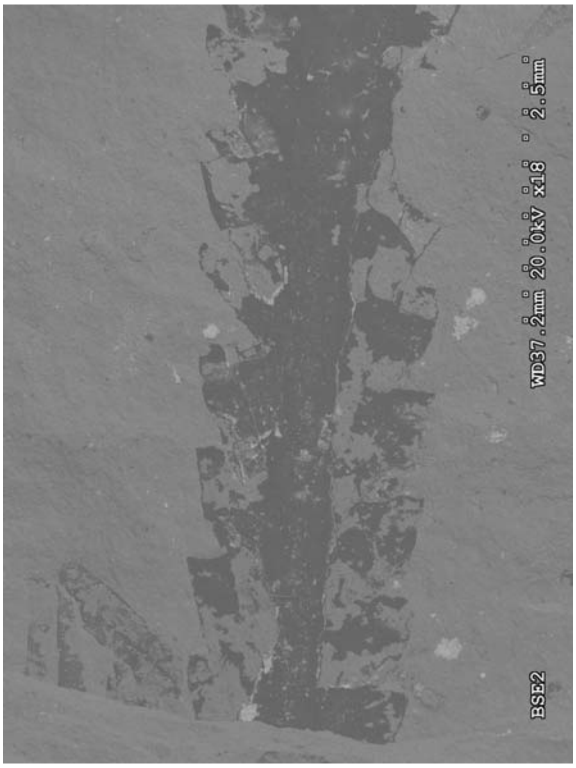
The 3-D visualization of the sample can be seen as a solid, showing the fissures with grains, or as a transparency, where the geometry of graptolites is manifested (Text-fig. 3). The accuracy of the µCT images was partly certified by the SEM observations of the sample surface. Text-fig. 5 shows the cleavage surface of sample 1. The µCT internal structure visualization of the shale core sample presents a flattened fragments of graptolites related to the cleavage surfaces and so sample 1 was cracked.

Furthermore, through µCT we obtained a visualization of grains (weak lamination) and quantification of graptolite and fissures volumes, as a parameter Vv, Which is defined as: Vv = Vf / Vs [-] where Vf is the volume of fissures as well as graptolite and Vs is the volume of the sample.

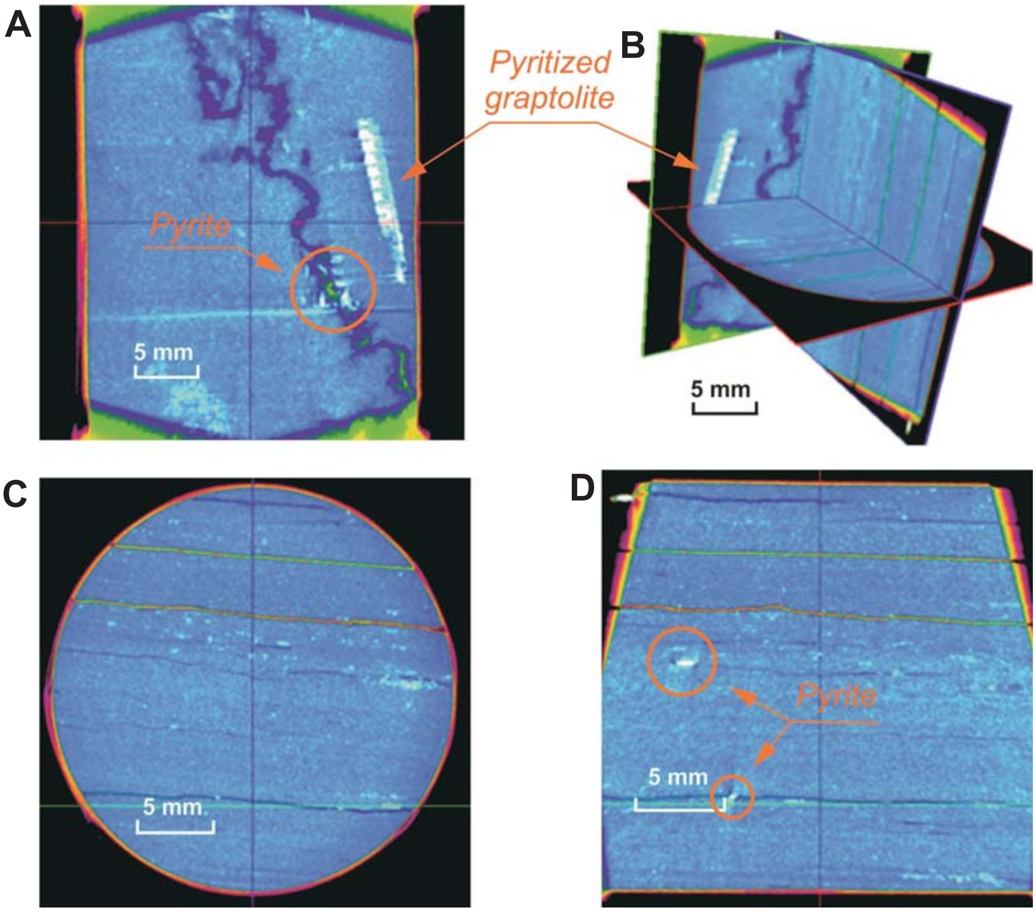
The 3-D numerical model of sample 1 is presented in Text-fig. 3A, B. It shows image segmentation (binarization) by interactive thresholding. Through such image division (value 0 – black pixels, value 1 – white pixels) the ratio of graptolite and fissure volume to the total volume of the rock sample is Vv = 2.8%. Compared to the previous 3-D model, the numerical model of sample 2 (Text-fig. 3C) shows multiple remains of graptolites. Furthermore, the ratio of graptolite and fissure volume to the total volume of sample 2 is Vv = 1.57%.

A magnified 3-D model of sample 2 is presented on Text-fig. 3D. The image is clear due to median and despeckling filters. It clearly shows some cross-sections of two monograptid graptolites.

The cross-sections through sample 1 in three perpendicular directions show weak lamination of the rock, pyritized graptolite and pyrite (Text-fig. 6), which may indicate anoxic conditions of sedimentation (Saupé and Vegas 1989; Roychoudhury *et al.* 2003; Zatoń *et al.* 2008). Weak lamination and pyrite were also observed in other samples. Pyrite is a high density mineral (circa 5 g/cm$^3$) and is seen as shining white spots, often surrounded by a streak of reduced absorption values areas (Cnudde 2005). SEM analysis of detected graptolites showed that they are partly pyritized and thus they shine intensely on µCT images.



Text-fig. 5. Topography SEM image of the sample 1 cleavage surface



Text-fig. 6. Sections of sample 1, depth 2871.40 m, Opalino core, in the region where a graptolite fossil was detected, due to the contrast of linear attenuation coefficients. A – Longitudinal section parallel to the height of the sample. B – Overview of all sections. C – Cross-section perpendicular to the height of the sample. D – Longitudinal section parallel to the height of the sample and rotated 90 degrees to section A

## DISCUSSION

The main contribution of this research is the demonstration of the applicability of X-ray computed microtomography for paleontology by visualization of the internal structure of shale reservoir rocks. Furthermore, the parameters required for µCT analysis of such samples were identified and the fossils with fissure content were determined, which is an important factor in the analysis of reservoir rocks. Based on the results obtained through binarization, 3-D numerical models of graptolites were obtained. In this study we used the thresholding method of binarization, which in the case of low diversity of material can be subjective (Beckers *et al.* 2014). Nevertheless, in the case of graptolite visualization it is a satisfactory method, showing key features of graptolites enabling determination of the species level. The µCT method differs from standard solutions, used in paleontology, due to the non-destructive character of analyzing the internal structure of the sample and the wide range of options available in terms of image processing and analysis.

Graptolite visualization was obtained during multi-stage tests of rock samples for characterization of potential hydrocarbons exploitation. The internal structure of the samples was analyzed by µCT and elastic parameters were determined using the ultrasound method (Kaczmarek *et al.* 2015). The last stage of the study was destructive compression and permeability tests, which determined the size and shape of samples to be cut from the rock core. In the case of complex geological conditions, the availability of samples and test length restrict the research possibilities. The µCT method can significantly accelerate and facilitate both industrial and scientific studies by determining several sample features at the same time. In order to obtain a resolution of a few µm for further analysis, after localization of graptolites in the sample the smaller fragments with graptolites should be cut out. After cutting out the smaller rock fragments of the sample, the optimal scanning parameters have to be determined once more in order to achieve adequate image resolution (Machado *et al.* 2014).

In this study we prove the usefulness of a new alternative method for detecting and recognizing graptolites. Although graptolites are often visible on the cleavage surface, in some cases no marks occur. In this case, or when there are difficulties with dissolution of rocks (e.g., marlstone), the µCT method may be the only method of identifying graptolites.

## CONCLUSIONS

Black shale samples were analyzed using X-ray computed microtomography (µCT), which produced images with spatial resolution of approximately 25 µm. These images were used to image Silurian graptolites that were not visible on the surface of the Opalino core. As a follow-up to the detection of graptolites, further microtomography analysis (visualizations and image operations) made it possible to locate them in core samples and calculate the ratio of graptolites and fissure to the total volume of the rock sample (Vv). Numerical models of graptolites reveal promising applications for paleontological research, and reservoir exploration and characterization. Hence, in future, images from µCT will be open to ever-widening horizons as new technologies become available. In the context of paleontological analysis and shale gas exploration µCT is a non-destructive, effective and highly-promising technique.

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