Investigations of elemental content, microporosity, and specific surface area of porous rocks using ion and X-ray microprobes

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INTRODUCTION

Determination of physical properties of porous geological materials is of great importance for oil industry. The knowledge of rock properties is usually obtained from porosity studies such as pore size distribution, specific surface area determination, and hydrodynamic permeability. This study describes determination of elemental composition and measurements of the particular physical properties of geological samples (porous sandstone rocks) by means of the nuclear and X-ray microprobes working at the Institute of Nuclear Physics, Polish Academy of Sciences (IFJ PAN) in Kraków, with the special emphasis on the computed microtomography (μCT) method. Measurements have been carried out in cooperation with Department of Geophysics, AGH University of Science and Technology (FGGEP AGH UST) in Kraków. Composition of sandstone rock samples (few millimeters diameter), extracted from a drill hole at 2679.6, 2741.4 and 2742.4 m depth have been investigated using the 2.2 MeV proton beam (PIXE technique). Next, measurements of the porosity and the specific surface area of pore space have been carried out using the X-ray beam and the μCT technique. Basing on tomographic data obtained with the high spatial resolution, simulations of the fluid dynamic in void space of porous media have been carried out. Lattice Boltzmann method in 3DQ19 geometrical model has been used in order to predict the hydraulic permeability of the media. In order to avoid viscosity-permeability dependency the multiple-relaxation-time model with half-way bounce back boundary conditions has been used. Computing power-consuming processing has been performed with the use of modern grid infrastructure [1].

X - RAY COMPUTED MICROTOMOGRAPHY

The μCT measurements of the porous sandstone rock samples have been carried out with the use of an experimental line of the multipurpose X-ray microprobe at the IFJ PAN. The line consists of an open type Hamamatsu L9191 X-ray tube with microfocusing down to about 2 μm, a high resolution X-ray sensitive CCD camera (Photonic Science 70 mm VHR), and a high precision rotary stage (Kohzu RA07A-W). Depending on the required X-ray energy, the Hamamatsu tube is used with Ti, Mo, Ag, or W targets. In this measurement Ag target ($E_{K\alpha}$=22.2 keV) has been used. A small focus size and short focus-to-object distance enable to obtain images of samples with high magnification and the resolution of the order of few μm. The μCT measurements are carried out using home developed codes combined with commercial software. Fig. 1 presents a schematic view of the μCT setup. The X-ray source equipped with a turbo-molecular pump is shown in Fig 2.

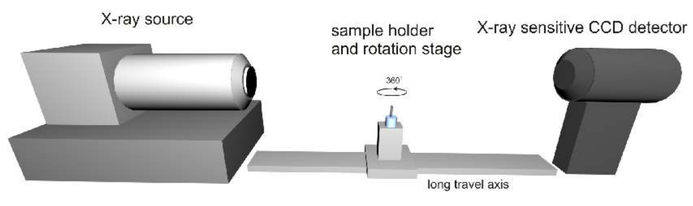


Fig. 1. Schematic view of the μCT experimental line

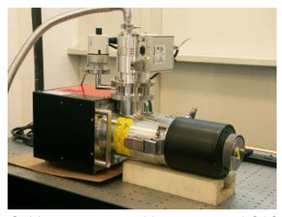


Fig. 2. X-ray source - Hamamatsu L9191 microfocusing X-ray tube.

PIXE MEASUREMENT

The determination of elemental concentration of trace elements has been carried out by means of the PIXE technique. A proton beam of 2.2 MeV delivered from the VdG accelerator (HVEC K-3000) at the IFJ PAN, Kraków has been used. The beam has been collimated down to approximately 1 mm^2 on the sample. The induced X-rays were registered by the CANBERRA Si(Li) detector with a resolution of 160 eV for the energy of 5.9 keV. The backscattered protons were registered by a silicon surface-active detector and used for X-ray spectra normalization. The acquisition time of characteristic X-ray spectrum was set to 600 s for each sample which corresponded to the charge of about 0.25 C. For quantitative determination of trace elements concentration the following external standards have been used:
IAEA-SD-N-1/2, IAEA-SL-1, IAEA-SOIL-7 [2]

Fig. 3. PIXE experimental line

Fig. 4. PIXE vacuum chamber

LATTICE BOLTZMANN METHOD

Lattice Boltzmann method (LBM) is a class of computational fluid dynamics (CFD) methods for simulation of fluid dynamic. It is a relatively new simulation technique for complex fluid systems which has attracted interest of computational physics researchers. In the LBM instead of solving the Navier–Stokes equations, the discrete Boltzmann equation is solved in order to simulate the flow of a Newtonian fluid with collision models such as Bhatnagar-Gross-Krook (BGK). Unlike the traditional CFD methods, which solve the conservation equations of macroscopic properties i.e. mass, momentum, and energy numerically, LBM models the fluid consisting of fictive particles, and such particles perform consecutive propagation and collision processes over a discrete lattice mesh. In this computations D3Q19 (3 dimensions, 19 discrete velocity vectors) model has been used (Fig. 5). Due to its particulate nature and local dynamics, LBM has several advantages over other conventional CFD methods, especially in dealing with complex boundaries which occur in porous media, as well as in parallelization of the algorithm. In order to avoid viscosity-permeability dependency the multiple-relaxation-time (MRT) model with half-way bounce back boundary conditions has been used.

CODE BENCHMARK

In order to check correctness and speed of the used code a test run simulating Poiseuille flow between two parallel plates has been carried out. The vertical velocity profile has been obtained and compared with analytical solution (Fig. 7). A good agreement of simulation results with analytical prediction was observed.

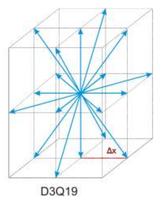


Fig. 5. Velocity vectors available in D3Q19 model

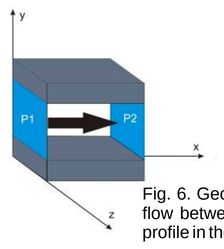


Fig. 6. Geometry of the Poiseuille flow - flow between two parallel plate. Velocity profile in the y direction is parabolic

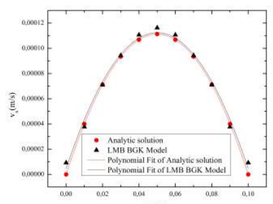


Fig. 7. Fluid *x* velocity component as a function of *y* coordinate. Simulation results vs analytical solution.

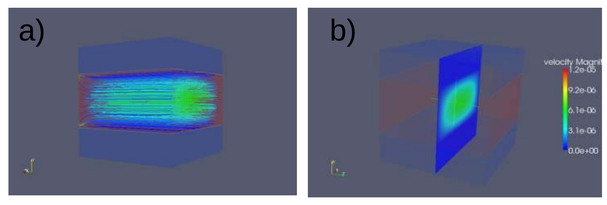


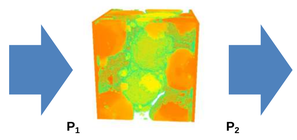
Fig. 8. Visualization of fluid velocity field. Streamlines a) and b) *x* velocity component. Arbitary simulation units have been used

MEDIA PERMEABILITY DETERMINATION

Simulations were performed on 255x255x255 lattice (voxel physical dimensions = 20 x 20 x 20 μm^3) with geometry of the medium obtained from μCT measurements and binary sampled into two regions – fluid and void space.
The permeability k of the medium was determined using the Darcy law:

$$< q_x > = \frac{k}{\mu}\frac{dP}{dx}$$

where:
P - pressure, <qx> - volumetric average of fluid flux, μ - dynamic viscosity.



RESULTS

All PIXE spectra were fitted with the well-established WinGupix software package (v2.1.4) [3]. Value of chi-squared varied from 2.12 - 10.7. Tab. 1 contains results of the trace elements concentration measurement in investigated samples, while fig. 9 presents the results in the chart form.

Depth [m]:	2679.6		2741.4		2742.4	
	[ppm]	± [ppm]	[ppm]	± [ppm]	[ppm]	± [ppm]
Al	180	20	370	30	60	5
P	< MDL		350	20	< MDL	
S	320	10	440	20	< MDL	
Cl	5000	50	3600	40	6600	40
K	20000	100	9800	65	6000	30
Ca	15500	200	2000	35	3200	20
Ti	3200	45	3000	40	740	10
V	< MDL		< MDL		80	10
Mn	350	50	< MDL		320	15
Fe	30000	200	11000	100	28700	100
Cu	230	20	260	20	120	20

Tab. 1 Results of the trace elements concentration measurement in investigated samples

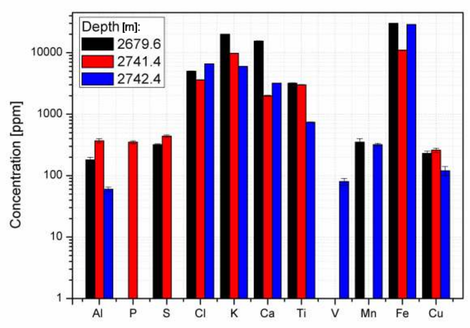


Fig. 9. Graphic representation of trace elements concentrations

An open source LBM solver „Palabos" [4] has been used in order to calculate total permeability of the media. The average permeability of the sample extracted from drill hole at 2679.6 m depth has been estimated to 20.0 ± 4.2 mD ($1D = 9.869233\times10^{-13}\ m^2$).

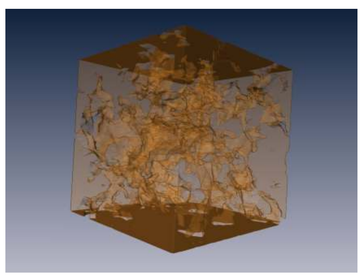


Fig. 10. 3D view of reconstructed sandstone microstructure

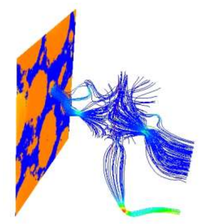
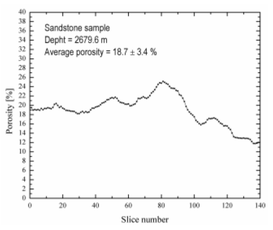


Fig. 11. An example of fluid velocity field obtained from LBM simulation (streamlines representation)

Depth:	Specific surface area	
[m]	[μm^{-1}]	±[μm^{-1}]
2741.4	0.25	0.01
2679.6	0.19	0.01

Tab. 2. Results of the specific surface area assessment



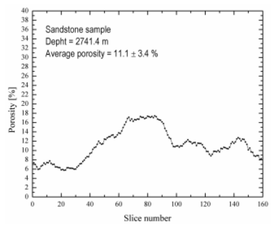
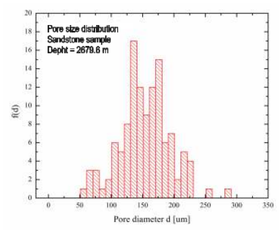


Fig. 12. Results of the porosity evaluation



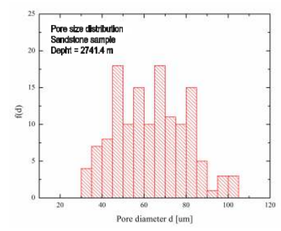


Fig. 13. Pore size distribution

The porosity (Fig. 12), specific surface area (a property of solids which measures the total surface area of porous media per unit of volume) (Tab. 2) and pore size distribution (Fig. 13) have been determined with the use of home-made code. Results of the porosity measurement are in reasonable agreement with results obtained with the use of the mercury porosimetry technique (15.28 % for the sample from 2679.6 m depth).

We would like to thankfully acknowledged:
- The Foundation for Polish Science and Technology (purchase of hardware and software for the X-ray microprobe, grant No. 222/FNiTP/119/2005)
- Ministry of Science and Higher Education of the Republic of Poland (grant No. DPN/N15/COST/2010)
- MP0601 COST Action - „ Short Wavelength Laboratory Sources"
- Support in the form of the DOCTUS scholarship cofounded by European Social Fund

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[3] http://pixe.physics.uoguelph.ca/gupix/main/
[4] http://www.lbmethod.org/palabos/

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