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MULTIPHASE FLOW EXPERIMENTS IN ORDER TO UNDERSTAND THE BEHAVIOUR OF (PARTLY) SATURATED COALS AS A GAS RESERVOIR: EXAMPLES

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(6 figures)

ABSTRACT. The use of coal seams as reservoirs implies that new ways of coal characterization are required. The storage capacity of coal has to be related to the ability of gases and fluid to migrate through a coal seam. Sorption and diffusion behaviour of the matrix and the cleat related Darcy permeability are the most important parameters for the determination of its reservoir properties. A new method for assessment of interfacial phenomena associated with carbon dioxide and methane transport/sorption processes in coals is presented. The article describes two innovative kinds of laboratory experiments. The flushing experiments characterize the flow properties of a solid coal core under simulated in-situ conditions. The pendant drop cell experiment characterizes the interactions of coal, water, carbon dioxide and methane. For that reason the equipment and experimental procedures of two flushing devices and the set-up of a transformed pendant-drop cell are explained. Besides procedures, technical difficulties, pitfalls and general interpretations are discussed in experimental examples. One of the major issues is, "how the experimental results are combined with known geo-parameters, in order to be used as input parameters at cleat and field scale".

Keywords: Coal, CO2-sequestration, Flushing Experiments, Pendant Drop Cell Experiments.

1. Introduction

Enhanced production of methane or sequestration of gases, such as carbon dioxide or flue gas, is practicable on condition that coal is considered both as a gas reservoir and as a storage medium. Normally the productivity of coal is measured by its drainage capacity of methane and water, as a function of pore pressure reduction. For conventional coal-gas production or coalbed methane production (CBM) this perception satisfies. However, in reservoir terms, considering high sweep efficiencies for both production and injection, the spatial characteristics of coal have to be related to its maximum internal accessibility at maceral scale at in-situ conditions. Special coal parameters such as cleat permeability, diffusivity and sorption are to be related to conventional clastic reservoir parameters, such as water/gas saturation, pore size distribution, capillarity, etc.

For this reason, coal cores are to be examined in "look alike" experiments. Hence, Delft University, Department of Applied Earth Sciences, is performing flushing experiments in high-pressure autoclaves. The cells usually contain cores, up to 1000 cm^3 in size, which are charged with methane and water. At pseudo in-situ conditions, they are flushed over months with CO2 and flue gas. The experiments provide information on volumetric changes and stress-dependent cleat permeability. In addition, the sorption/desorption behaviour of the coal matrix for methane, carbon dioxide, other gases and water are examined.

In the course of this study, it became clear that the carbon dioxide/methane exchange ratios, differed considerably with changing water saturation. For this reason a Pendant Drop Cell was adapted to in-situ coal conditions. In this vessel, it is possible to study the solid-liquid-gas interactions in a changing coal-water-carbon dioxide-methane system. CO_2 droplets on a water-coal interface are visualized and observed at increasing fluid pressures. The images give information about the CO_2 dissolution in water and its diffusion into the coal.

2. Flushing experiments

Flushing experiments in autoclaves, as performed in many experiments by Wolf et al. (1999), de Haan (1999) and Bertheux (2000), are in use to obtain data related to:

- Stress-dependent cleat permeability.
- Langmuir type sorption isotherms for carbon dioxide and methane.
- Displacement of water and methane by carbon dioxide.
- Displaced volume in relation to time, temperature, pressure and expansion or shrinkage.

2.1. Equipment

Two autoclaves have been constructed to flush methane from (water saturated) coal cores. Figure 1 shows an outline of the experimental set-up, which basically consists of a high-pressure reactor with a coal core. To apply the compacting stress to the coal sample, it is placed in a flexible synthetic core holder, which is compressed with an annular pressure by oil or nitrogen. The maximum annular gas pressure is 110 bar; the maximum oil pressure 500 bar. The maximum length and diameter of the sample are respectively 800 mm and 80 mm. Generally, the samples have a length varying from 200 to 300 mm and a diameter from 72 to 78 mm. For an optimal gas injection and axial compaction, the rubber sleeve contains at both ends of the specimen a ceramic end-piece. The end-pieces are connected with a fixed and a movable steel tubing. A storage vessel and a gas booster (ISCO-pump) are used to inject various gases and water into the coal at minimum injection flow rates from 0.7 cm^3/hr. In the regular tests, the maximum pore pressure is 10 bar below the annular pressure. At the production side the tubing is connected with a back valve reducer (safety), after which a fluid capture container is placed on a balance. Here the amount of produced water is measured. Then a flow analyser and chromatograph are connected to measure the flow rate and gas composition. To measure the volumetric changes, the movable exhaust tubing of the coal core is connected to (axial) displacement transducers. During the experiments the compaction pressure, inlet/outlet pressure and differential pressure over the sample, displacement, and temperature are measured every minute. Since an experiment takes three to eight weeks, an automatic registration of all environmental parameters is essential. During an experiment, the pressure and displacement readings are used to control the volumes of (partly) flushed coal, gases and fluids against time and according to the requested values for a specific p,v,T,t-environment. The results are used for the construction of a mass balance, a volume balance and related sweep efficiencies.

2.2. Experimental procedure

During the initialisation phase the sample is inserted in the autoclave and the tubing system is tested with helium for leaks. The sample is attached to a vacuum pump for at least twenty-four hours up to one week, to remove the (adsorbed) gases and water. Thereafter the coal is filled in several cycles with methane, until the required pore pressure is reached (Fig. 2). After each injection cycle, the methane is allowed to adsorb onto the coal matrix, until a near equilibrium is reached for the specific pressure and temperature. The injected methane is counted by a mass flow meter. During the charging phase, the autoclave is brought to the desired temperature. Depending on the type of experiment (dry or water saturated coal), water is injected and both, the injection reservoir and the pump are brought to the same pressure and temperature conditions as the methane-filled sample. When, during the previously described procedure, the injection pressure reaches the maximum methane filling capacity of the coal sample, one needs to be careful with an expected or a calculated highest amount. Overfilling easily induces sample fracturing of the coal, displacement of an end-piece, which creates a methane-filled void, and finally rupturing of the sleeve. Maximum methane contents usually are reaching up to 30 m^3/ton of coal. To meet sub-surface injection conditions with effective permeabilities, the difference between the annular pressure and the pore pressure is kept at 10 to 20 bars. When CO_2 injection starts, the gas analyser determines the relative amounts of CO_2, CH_4, N_2 and other gases in the product gas. The N_2-detection

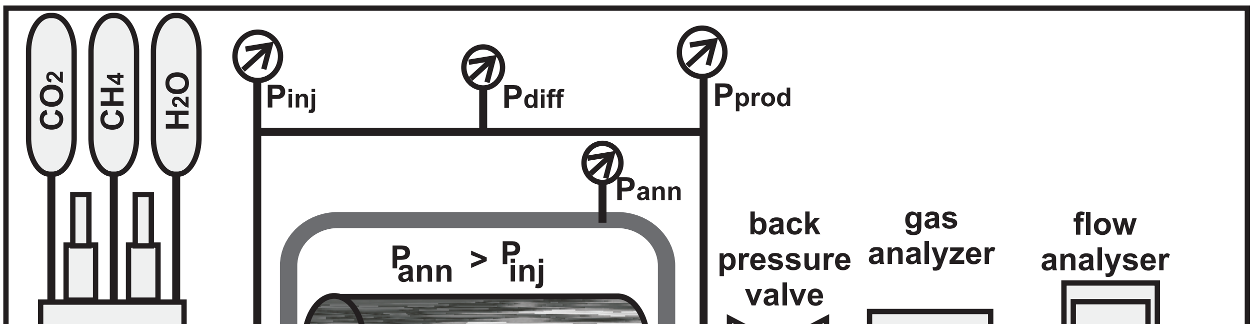

Figure 1. General scheme of a laboratory setting for flushing of methane with industrial gases out of (water) saturated coals. Revised after Wolf et al. (2000).

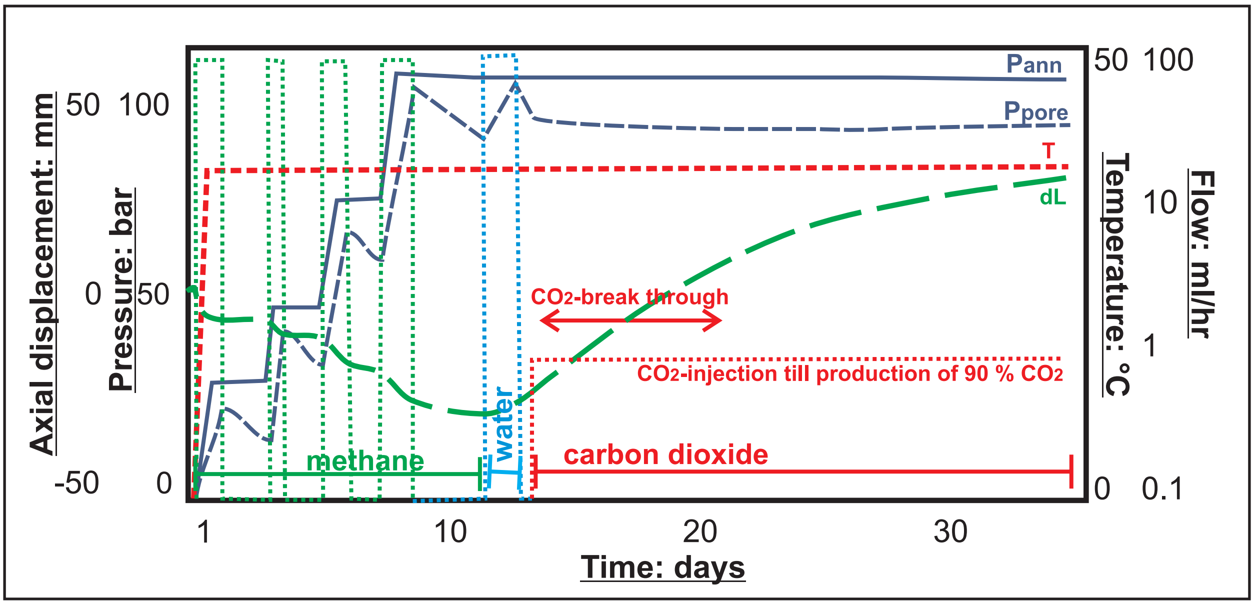

Figure 2. An experiment from the moment of methane injection. Pressure, temperature, injection/production, flow and axial displacement versus time.

is used in the 110 bar vessel for leak control. During the tests, the recorded data serve as an iterative feedback in order to rule out environmental influences in the interpretation afterwards. These procedures are extensively described by Wolf et al. (1999), de Haan (1999) and Bertheux (2000).

2.3. Experimental results

Figure 3 is an example of the course of an experiment, including the cumulative flushing results. A flushing experiment can be compared with a transition zone in a coal seam, going from a virgin reservoir into a (partly) flushed reservoir. Hence, all stages of the experiment are used for the reconstruction of the sample physical properties. The results are an input for reservoir modelling work.

2.3.1. Nitrogen flushing and methane charging

Cleaning a core by flushing with nitrogen, provides information on its cleat permeability. Flushing tests at different annular stresses up to 20 bar pressure difference (confining pressure minus pore pressure) show in general effective cleat permeabilities higher than 0.1 mD. Evacuation of the sample and stepwise isothermal charging with methane to a required pore pressure, provides information on CH_4 sorption, that can be evaluated in terms of a Langmuir isotherm (Fig. 3A; CH_4 injection). During charging, after each pressure step, the pore pressure drops slowly. This pressure reduction provides information regarding the bulk diffusion rate and the bulk CH_4 sorption for a specific coal sample, on a specific pressure and temperature.

2.3.2. CO_2-flushing

CO_2-flushing, or replacement of methane and water by carbon dioxide, provides information on displaced volumes in relation to time, temperature, pressure and volumetric changes of the core (Fig. 3B). Knowing the amounts of originally injected methane and water, the amounts of produced fluids and gas, and the composition of the gas (Fig. 3D), it is possible to calculate the displaced volumes of methane, carbon dioxide and water (Fig. 3C). The resulting net volumes injected and produced give the sweep efficiencies for all migrated components (Fig. 3E). Sweep efficiency is defined as the ratio of the produced amount and the amount of gas or fluid originally present in a porous medium.

With the known sample dimensions and sweep characteristics, one can estimate the extension of a transition zone for these cores, under the specified pseudo in-situ conditions. The transition zone is the distance between the virgin rock, with the original amounts of gas and fluid in its porous system and the rock in which a maximum of the original fluid and gas has been replaced by an injected medium. The first experimental series prove that high injection rates create large transition zones. Fast CO_2-transport through the cleat zones is associated with a "delayed" effect of diffusion and sorption into the coal matrix. Results on the "displaced volume" values at low injection rates (< 1 cm^3 of CO_2/hr) show that, at 80 % CH_4 sweep efficiency, a minimum transition zone of 4 meters can be expected. In addition, it is recognized that dry coals have the highest methane sweep efficiencies and the narrowest transition zones.

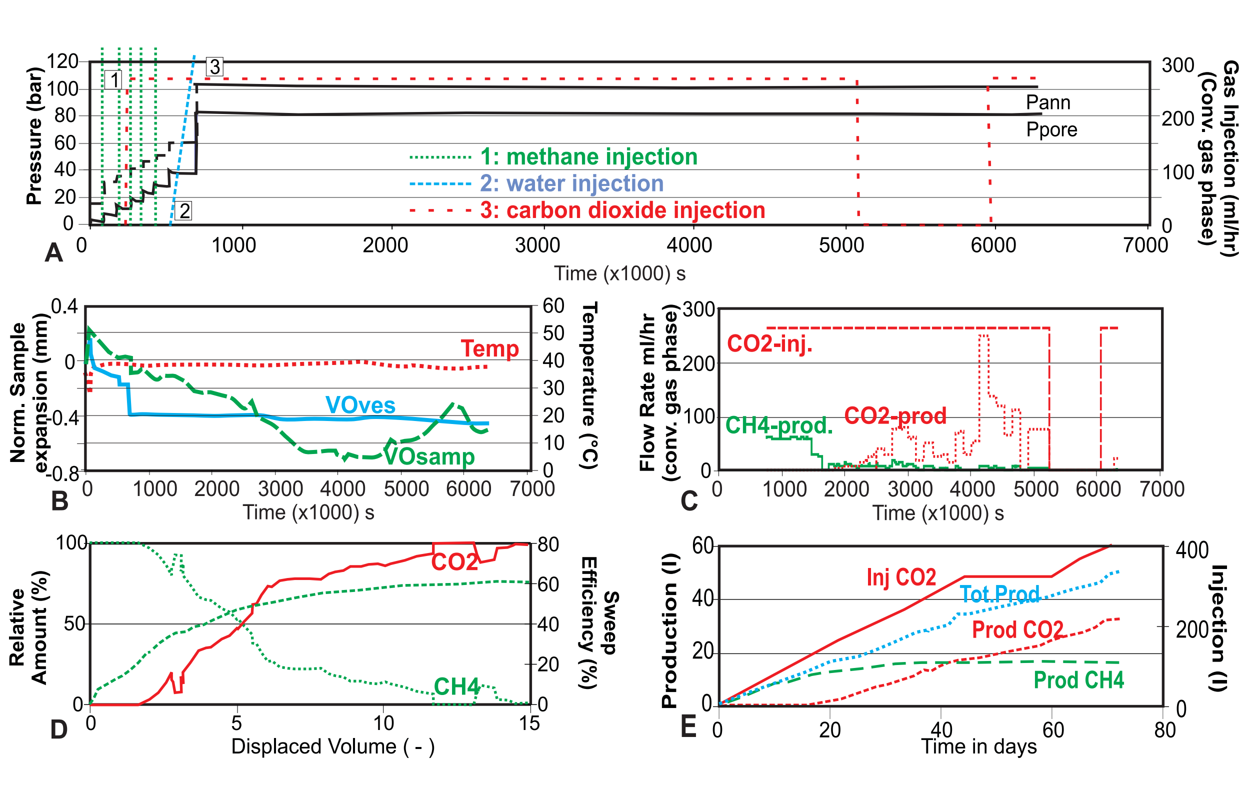

Figure 3. Example of data-acquisition of a water free flushing experiment. A: annular pressure and pore pressure versus time, including the different injection phases, B: temperature and axial displacements (strain determination) of the vessel (VO_{ves}) and sample (VO_{samp}), C: injection and production history of methane and carbon dioxide, D: produced gas and methane sweep efficiency versus displaced volume, E: cumulative injection and production versus time.

2.3.3. Displacement transducers

Increase of annular pressure and temperature cause an expansion of the pressure vessel at the end pieces and a hydrostatic decrease of the coal volume. In addition, injection of methane and water during the initial phase and carbon dioxide during the flushing phase will increase the core volume. Hence, two displacement transducers measure the change in axial length of the coal core (VOSAMP) and of the pressure vessel (VOVES). This change in volume, associated with CH_4-, water and CO_2-injection, gives the pressure-, temperature- and time-related dimensional effect of different gases on coal. In this way, a mass balance of the experiment is supported by a volume balance. In the experiments large volume increases are recognized, related to CO_2-injection (Fig. 3B). However, based on this volume balance, the currently accepted relation between coal swelling by CO_2-charging and a related serious permeability reduction (Pekot et al., 2002) could not be confirmed.

3. Pendant drop cell experiments

Carbon dioxide in a coal-methane-water system is known to act as a good displacement fluid for the methane-water mixture. In addition, the flushing experiments demonstrate that the methane sweep efficiency of a dry coal is high compared to the efficiency of a water-saturated coal. Therefore, the water saturation of coal is important for the transport capacity of a cleat system when carbon dioxide and methane are migrating. When considering a coal in terms of a siliciclastic reservoir, the system could be considered as water wet or (instead of oil wet) CO_2 wet. The clastic pore space represents the complement of the mineral matter present. In coal the matrix complement, i.e. the cleat system, is the major pore system for transportation. The major differences with respect to a hydrocarbon reservoir rock are; the absence of oil, the presence of methane and the major "second" matrix-pore system at the micro/nano-size scale. Due to the different nature of the matrix matter, cleat system and pore filling, the following questions are put forward:

- To which extent is it possible to determine the wettability of a coal sample? Is it possible to refer to a cleat-matrix system as water wet, methane wet or carbon dioxide wet?
- Will there be an effect of the maceral type on the degree of wettability and in consequence, on sweep efficiencies?
- Can diffusion and sorption characteristics, of methane and carbon dioxide droplets on a coal face, be reconstructed from droplet-images?

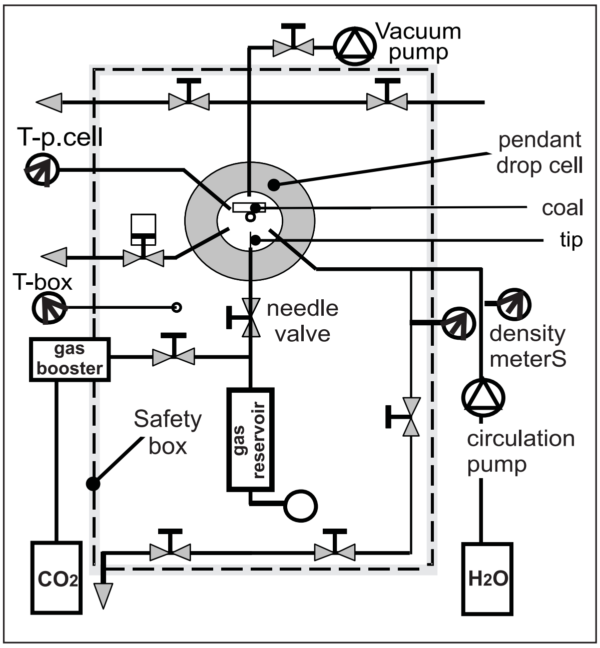

Figure 4. Scheme of the experimental setting. The cell is placed perpendicular to the situation on the photograph in fig. 5.

- Can a volume change of the coal matrix by sorption/ desorption be visualized?

To answer these questions, a conventional pendant drop cell was reconfigured in order to visualize and record these effects. In-situ p,T-conditions are imposed on a coal interface - water - gas (methane or carbon dioxide) system. The initial experiments show that it is possible to observe droplets at a coal interface and register volume changes and contact angles, through time and at different fluid pressures.

3.1. Equipment

A pendant drop cell, as shown in Figures 4 and 5, consists of a high-pressure cell with inlets and outlets for both the liquid component and the gas component. The fluid pressure in the cell is controlled by a pump. A gas-booster and reservoir are connected through a needle valve with a small tube in the cell, to produce controlled gas droplets (Fig. 4). The cell is placed in a safety box, which also keeps the cell at a constant temperature (Fig. 5). To keep the process as constant as possible, all pressures and temperatures in the system are monitored. For our purposes the inner-cell was rotated by 180º, which changed the conventional system in two ways:

1. A coal sample is placed in the upper part of the cell and "catch" a droplet, which rises from the tube tip below (Fig. 4).
2. The glass windows, installed for monitoring the event, transmit monochromatic light. This prevents image distortion by refraction.

An endoscope, connected with a web cam, is placed in front of one window, to monitor the behaviour of a gas droplet at the coal surface (Fig. 5). The images gathered are processed by image analysis to determine spatial attributes. In this way, different combinations of gases, fluids and coal types are studied under pseudo in-situ "pore" conditions.

3.2. Experimental procedure

At first the pressures, temperatures, fluids and gases are defined. During injection and pressurizing the cell, the fluid is slowly circulated through the system for two reasons:

1. A heater installed on the reservoir and tubing pre-heats the fluid to the same specified temperature, as present in the cell.
2. Fluids in the cell are contaminated with the injected gases. They have to be replaced after each droplet experiment.

Injection of a gas droplet occurs through a needle valve-controlled tube. The droplets are generated at low and specified rates and sent to the flat coal surface.

Two types of experiments can be carried out:

1. An injected droplet is monitored during fluid pressure increase at the fluid-coal interface.
2. At different fluid pressure steps and temperatures, the gas droplet behaviour at the coal-fluid interface is monitored.

With these experiments, it is possible to observe the shape of a droplet at changing fluid pressures. The images are subsequently processed by image analysis, with respect to the spatial characteristics; area, width, length, perimeter and contact-angle (Fig. 6). Calibration and centration of the endoscope and web cam, including the focusing on the droplet, are essential activities, which have to be carried out thoroughly. Blurred images are difficult to use or even unsuitable for use with image analysis.

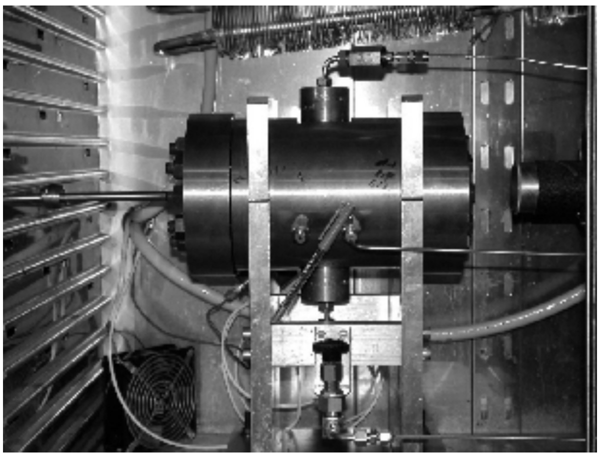

Figure 5. For coal – water - gas interaction, a reconstructed pendant drop cell with an endoscope (left centre).

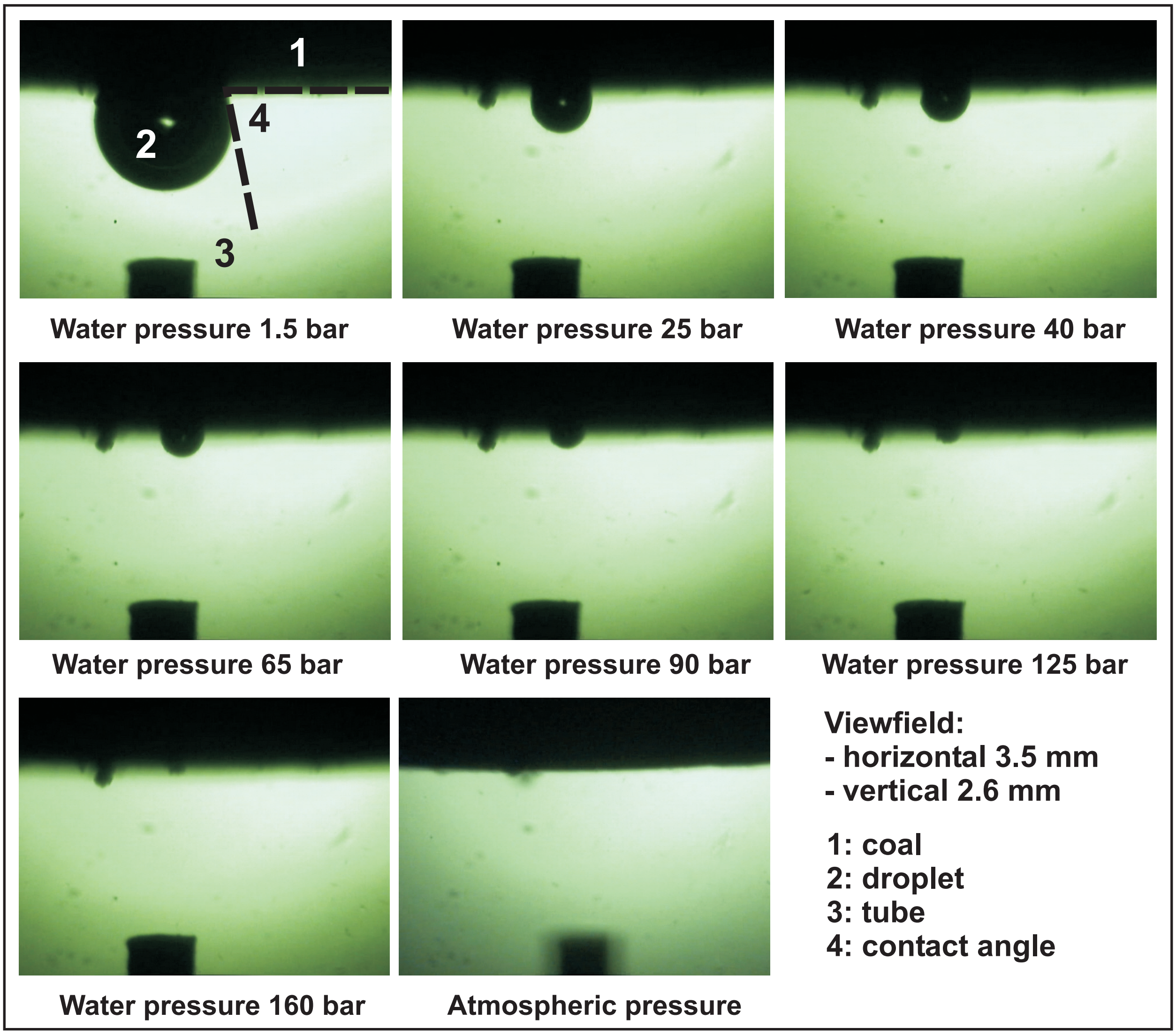

Figure 6. CO_2-droplet at a coal face. Notice the volume reduction at increasing fluid pressure and disappearance into the coal matrix at pressure reduction.

3.3. First results

Up to now, most of the work involved the fine-tuning of the apparatus on constant flow, pressures, temperatures and accurate image acquisition. A few pressure experiments have been carried out and images acquired. As shown in figure 6, the images have to be enhanced in terms of sharpness. Figure 6 also shows that droplets do not necessarily move straight upward. As a result, the endoscope needs to be flexible to be able to "catch" a sharp image of a droplet and a sharp coalface. Image analysis on the droplets has not yet been performed, since picture enhancement is needed.

Some testing series (Figure 6), show serious effects of water and the coal matrix on a CO_2-bubble. At fluid pressure increase, the bubble volume reduces. If CO_2 does not react with the fluid or coal, volume decrease should be a reversible process. However, with pressure reduction the bubble does not increase; migration into the coal matrix and into the water is the most likely explanation for this observation. These results show the interaction of gases with the pore fluid and coal matrix. The contact angle between the gases and the coal surface point to the wettability of pre-defined gas/fluid/coal-matrix systems.

The experiments will be extended to bubble behaviour in (partly) water saturated and methane saturated coals. In addition, attention will be focused on water salinity and gas compositions (i.e. flue gas).

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