VISUALIZATION OF ROCK SAMPLES IN THEIR NATURAL STATE USING ENVIRONMENTAL SCANNING ELECTRON MICROSCOPE.

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Abstract

Among the new tools which have been developped recently in the area of rock characterization at the microscale level, Environmental Scanning Electron Microscope (ESEM) is particularly relevant in petroleum technology. This technique allows visualization of rock samples in their natural state in presence of fluid distribution, and during microdynamic experiments in presence of both brine and oil. In addition to this dynamical process recording, imaging and elementary analysis are also available.

This new technique has been used in our laboratories to perform various experiments on gas/liquid and liquid/liquid systems, on core samples coming from different reservoirs known for their wettability behavior.

First, a set of condensation-imbibition tests on shaly samples were carried out to investigate the anticipated preferential affinity for water of illite versus kaolinite. This direct visualization of contact angles and imbibition effectiveness in an air/brine system showed a clear affinity of clays for brine and an imbibition process mainly controlled by shape factors.

Secondly, a step by step visualization of fluids distribution in an oil/brine system during initial water saturation or residual oil saturation installation has been performed. This allowed to visualize oil and brine distribution in the case of three reservoir core samples, one water wet, one intermediate wet and one oil wet. A series of images illustrates some of the oil/brine displacement mechanisms at the microscale.

Introduction

Wettability has always been considered as one of the main parameters in controlling fluid distribution, and two phase flow behavior. Many authors have already described these phenomena very accurately using various methods (ref.1). For example, correlation between wettability and capillary pressure as well as relative permeabilities (ref. 2, 3), contact angle determination and correlation to wettability (ref. 1, 3, 4), fluid displacement mechanisms versus wettability (ref. 3, 4, 5, 6), have been studied extensively. However, the microscale description of wettability is, to our knowledge, more recent and progresses significantly with the actual techniques of visualization (ref. 7).

The aims of this study were first to show contact angles in a porous medium in its real state, and not on an synthetic substrat, and secondly, to explain some of the oil/brine displacement behavior at microscale.

These visualizations allowed a better understanding of some of the microscale phenomena involved in rock/oil/brine systems, which are especially important in case of intermediate or oil wet reservoir : thin film flow and their efficiency, clear affinity for oil or brine, influence of local mineral heterogeneity on fluid distribution.

For the purpose of this study, Environmental Scanning Electron Microscope (ESEM) was used to perform observations. One of the main advantages of this technique is to allow visualization of fluids in a porous medium without any complicated sample preparation such as metal coating for Scanning Electron Microscope (SEM) or freezing for Cryo-Microscopy (Cryo-SEM) for example. To summarize, ESEM has all the advantages and performances available in SEM, but in addition, microdisplacement, condensation or imbibition with a direct visualization can be performed.

ESEM description

The ESEM technique allows visualization of liquids and natural wet samples (i.e. reservoir rocks with both fluids) thanks to its following characteristics :

- it operates with a differential pumping system which creates a pressure gradient in the electron optical column (fig. 1). Several chambers seperated by a series of small apertures along the beam path allow to keep the electron beam source and the upper column under high vacuum, while the specimen chamber can be under pressure from 2 to 7 kPa.

- a new secondary detection electron system operating at chamber pressure (2 to 7 kPa) was used. Gas environment, temperature and pressure can be set up in the specimen chamber. For example, gas environment can be ambient gas, nitrogen, argon, water vapor, organic solvent vapor, etc ... Danilatos (ref. 8) developped the formulae governing gas dynamic and beam scattering in air. He showed that the spatial resolution of unscattered beam fraction can still be used to form an image with a good resolution as the one obtained under high vacuum.

Whence, ESEM incorporates the best features of conventional SEM (high resolution, depth of field, electronic signals detection, elementary analysis with X-rays detectors) with, in addition, the flexibility and easiness of use of a light microscope. It can be used to visualize and record in real time dynamical processes such as drying, melting, dissolution, cristallisation and chemical reaction at temperature up to 1500 °C. Some of these new possibilities are particularly relevant for petroleum industry, as it has been described by Mehta (ref. 7)

Environmental Secondary Detection System

As a conventional SEM, the specimen electron beam interactions produce electronic signals including secondary electrons, backscattered electrons and X-rays. The standard Everhart-Thornley detector cannot be used because of the high voltage requirement on its scintillator surface which would create electrical breakdown in gaseous environment. So, ESEM uses the Environmental Secondary Detection System first described by Danilatos (ref. 9). This new system involves the gas environment itself in the signal amplification process (fig. 2). Gas ionisation is induced by electrical field forcing collision between highly mobile secondary electrons and neutral gas molecules. Successive collisions liberate additionnal free electrons resulting in the cascading multiplication of secondary signal. The negative charges build up on

the sample are dissipated by the free positive ions created into the gaseous environment of the specimen chamber.

Experimental procedure

The ESEM (Electroscan Corporation) operates at various high voltages using the environmental secondary detector (EDS). Samples were :

- examined without any preparation or coating,

- visualized at various temperature (between 5 and 15 °C) and pressure (between 400 and 535 Pa),

- maintained in their natural state by controlling the specimen temperature with a Peltier thermoelectric stage. The chamber vapor pressure, controlled according to pressure and temperature relationship, allowed to keep the wet specimens fully hydrated during examination,

- tilted 18° toward the detector during the X-rays spectra acquisition.

The X-rays generated from the samples were processed with Noran instrument's work station (EDS2I). This system was equipped with a diamond window light element detector. The working distance was adjusted to avoid the parasitic detection X-rays of gold and copper of environmental secondary detector located above the sample.

Experimental description

Rock samples

First, a set of dynamical condensation/imbibition experiments was carried out on a reservoir core sample (rock A, fig. 3) containing both illite and kaolinite. Several authors (ref. 10) already worked on this reservoir using Cryo-SEM. They showed a preferential affinity for brine of illite versus kaolinite when brine and oil are present in the sample.

Secondly, and for illustrating our purpose on fluids/rock interactions in the most complete way possible, three different core samples were choosen in three different reservoirs of known wettability. This allowed to check the fluid repartition at various saturation states in a water wet, intermediate wet and oil wet conditions and verify the usual assumptions.

Considering the ESEM specimen chamber geometry, the core samples subjected to visualization had the following dimensions : 2.5 cm in diameter and 5 mm thickness. Also, to visualize the most representative surface, prior to ESEM experiments, the disks were slightly fracturated under saturating fluids conditions. Their characteristics are described bellow.

Rock 1 : Water wet case (fig. 6)

This sample is a sandstone mainly composed of quartz (76.5 %), mica and feldspars (9%), clays (14 %, mainly kaolinite). The total porosity is 27.9 %PV.

Two set of Amott-IFP tests (ref. 3) were carried out on this sample. The first one was performed at laboratory conditions after cleaning by solvents circulation to identify the mineral contribution in wettability. The second was performed after wettability restoration (aging period in stock tank oil) at reservoir conditions to obtained the usual wettability index, and the contribution of oil. The wettability index obtained was WI = 0.8, which indicates a clear water wet behavior.

Rock 2 : Intermediate case (fig. 7)

This sample is a fine to medium sandstone, mainly composed of quartz (95 %), feldspars (2 %), some heavy mineral traces, few micas, some siderite around quartz and feldspars, and some traces of pyrite.

Several waterfloodings at laboratory and reservoir conditions were carried out on this rock type. The relative permeabilities and capillary pressure determinations indicated a clear intermediate wettability behavior.

Rock 3 : Oil wet case (fig. 8)

This sample is mainly composed of quartz (79 %), dolomite (12.3 %), feldspars (6.2 %), clays (1.2 %) and some heavy minerals. The total porosity is 19 %PV.

As for Rock 1, a serie of Amott-IFP tests were performed before and after wettability restoration. The wettability index obtained was WI = -0.7, indicating an oil wet behavior.

Fluids

The fluids used were recombined brine (20 g/l NaCl) and neutral oil (Marcol 52, 11.5 cPo at 20°C), as the use of stock tank oil in ESEM conducts to a poor image quality. However, no tracer was used to improve image quality, and fluids were in their natural state (no freezing and no metal coating). Direct visualization allowed to identify oil and brine without any problems, the contrast between the two phases being important.

Saturation procedure

The samples were cleaned by solvents circulation, dried and then fully saturated in brine under vaccum. Inital water saturation (Swi) and residual oil saturation (Sor) states were obtained using the centrifuge technique. This procedure has been previously used by several authors (ref. 11), and was shown to be accurate and reproducible.

Results and discussion

Condensation-imbibition (air/brine) (fig. 3 to 5)

The experiment started with a cleaned and dried sample (rock A, fig. 3), containing mainly kaolinite (top-right), illite (bottom-left) and quartz (center). Then, pressure and temperature conditions were modified in

the ESEM chamber to induce a water condensation process inside the porous medium. Pictures were taken at various stages of this process (fig. 4 and 5) showing the following points :

- water invades first the kaolinite network,

- water droplets appear on quartz grain with increasing contact angles versus water saturation,

- a small amount of water is adsorbed on the illite agregate surface, leading to the progressive invasion of the network.

In the air/brine system, it appears that the wetting behavior is mainly controlled by shape factors. No major well defined intrinsic affinity for water of illite or kaolinite has been observed.

Rock samples fully saturated by water (fig. 9 to 11)

Full water saturation corresponds to the first step of the fluids installation procedure. No major differences between the three core samples were observed (fig. 9 to 11).

At this stage, water desaturation can be conducted under ESEM visualization to examine dynamically water removal from minerals.

Fluid distribution at Swi (fig. 12 to 15)

The initial water saturation set up was simultaneously conducted on the three samples using the centrifuge technique. Direct visualization by ESEM (fig. 12 to 14) showed significant contrasts between oil (deep black) and brine (light grey) allowing convenient identification of phases. In addition, this was checked by X-ray analysis, pointing out each phase and measuring the carbon/chloride ratio. A low carbon/chloride ratio indicates the presence of brine, and conversely, a high ratio allows to identify the presence of oil.

The morphological water repartition appeared to be mainly different for the three core samples :

- rock 1 (fig. 12) : a thin water film (about 2 or 3 μm thick) was spreaded on the pore surface, allowing only few contacts between oil and rock surface.

- rock 2 (fig. 13) : two behavior coexist in this core sample, one showing trapped water droplets (about 5 μm diameter) surrounded by oil (top-middle), the second showing a partial spreading of water on the surface (middle-bottom).

- rock 3 (fig. 14) : water appeared only present on the form of droplets (about 5 μm diameter), either trapped in the middle of oil, or in contact with the mineral with a very strong contact angle.

In addition, since the core sample was at Swi, it was possible to condensate water inside the porous medium, recording dynamicaly the process. Condensated water enlarged where the initial water was located (fig. 15), hence providing a way of looking at displacement of oil by water at the microscale level.

It is shown that initial water saturation has a very close dependency on wettability. Even if Swi is identical at the core scale in various wettability cases, the microscale fluid repartition appears completely different. This implies, that for given Swi, oil/brine/rock interactions vary, which has a direct effect on aging or waterflooding.

Fluid distribution at Sor (fig. 16 to 18)

The last step of this study was the visualization of the fluid distribution at residual oil saturation. As for initial water saturation, Sor was obtained simultaneously on the three core samples using the centrifuge technique.

As expected from the differences observed at Swi, the microscale fluid distribution at Sor depends a lot on the wettability. The three cases exhibited the following behavior :

- rock 1 (fig. 16) : oil is trapped as droplets (about 5 to 10 μm in diameter) surrounded by water, pore surface being always in contact with water.

- rock 2 (fig. 17) : the residual oil is on the form of droplets, in contact with both water and pore surface. The interface between oil and water is flat, and the contact angles between oil and mineral surface illustrate an intermediate wettability.

- rock 3 (fig. 18) : a thin film of oil (about 5 to 10 μm thick) is in contact with the major part of the pore surface. Water is in a continuous phase in the middle of the pore network with only few contacts with the minerals.

The microscale residual oil saturation repartition is strongly dependent on the wettability. As it has been shown by several authors, the recovery process is related to the phase repartition on porous media and film flow processes.

Conclusions

It has been demonstrated that Environmental Scanning Electron Microscope is a very useful and powerful tool which allows to visualize complex phenomena. Visualize porous media in their natural state in presence of fluids distribution and perform microdynamic displacements with both oil and brine are two main improvements in understanding oil/brine/rock interactions.

In the air/brine system, a clear affinity for water of illite or kaolinite has not been observed during dynamical condensation. The wetting behavior is mainly controlled by shape factors.

In the oil/brine system, it has been shown that the microscale fluids distributions are related to wettability. The macroscale (core scale) values of Swi or Sor can be the same in various wettability cases, but the oil/brine/rock distribution appears to be completely different. This has a strong influence on oil/brine displacements and should not be neglected.

Also, it has been clearly shown that ESEM is an accurate technique to visualize thin films and study their evolution versus fluids saturation.

Aknowledgements

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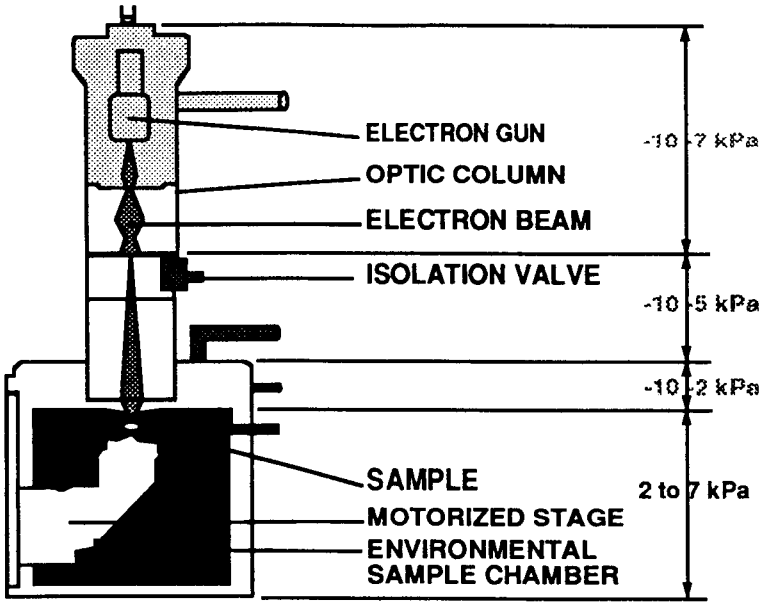

Fig. 1 - Shematic diagram of the ESEM

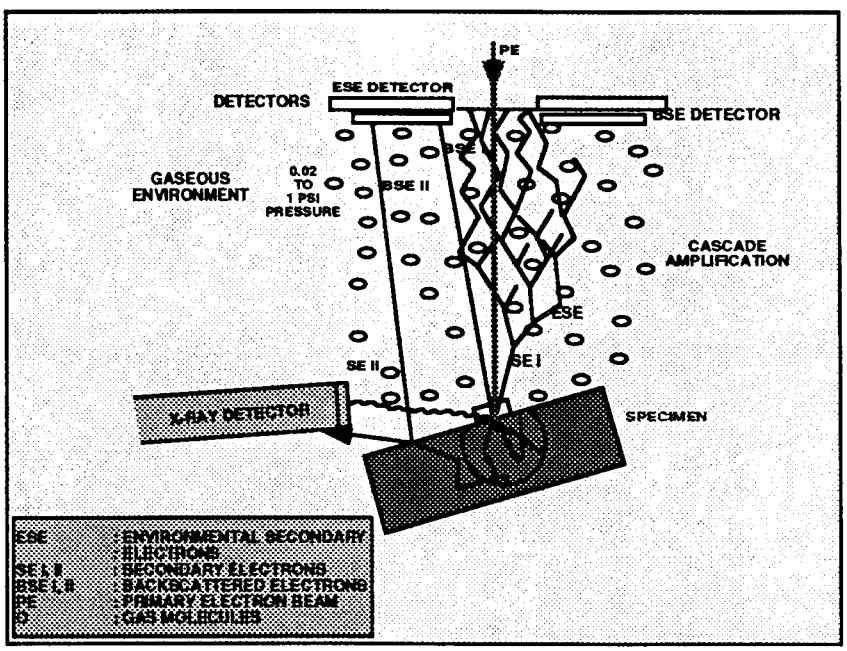

Fig. 2 - Schematic view of electronic signals generation and collection in the chamber of the ESEM

Fig . 3 - RESERVOIR CORE SAMPLE A CONTAINING ILLITE AND KAOLINITE

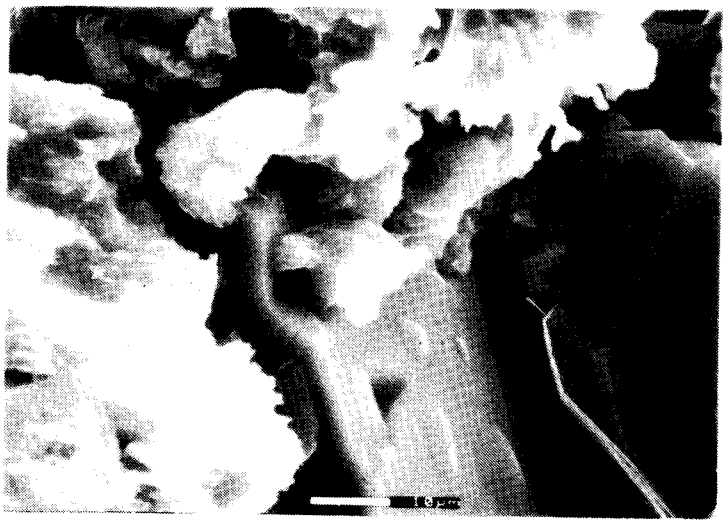

Fig .4 - INITIAL STAGE OF THE CONDENSATION/ IMBIBITION PROCESS (ROCK A)

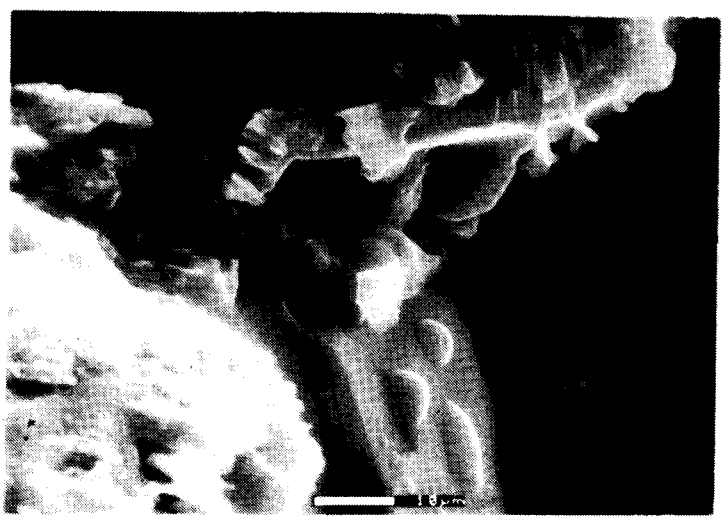

Fig .5 - INTERMEDIATE STAGE OF THE CONDENSATION / IMBIBITION PROCESS (ROCK A)

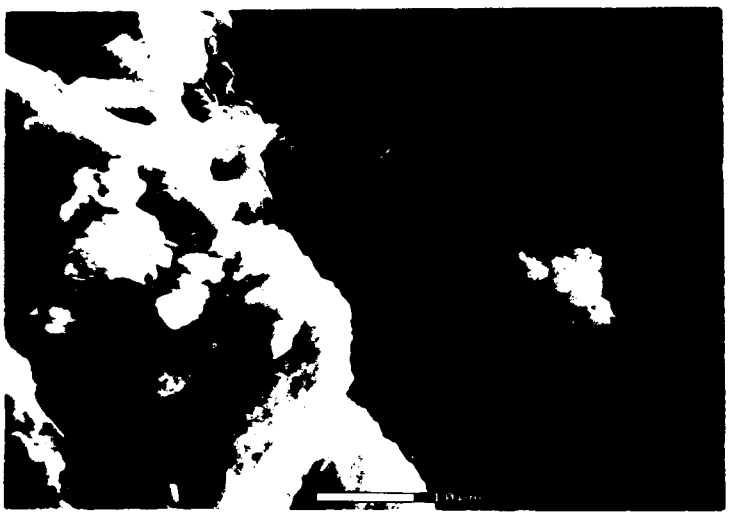

Fig . 6 - RESERVOIR 1
CORE SAMPLE
(DRY SPECIMEN)

Fig .7 - RESERVOIR 2
CORE SAMPLE
(DRY SPECIMEN)

Fig .8 - RESERVOIR 3
CORE SAMPLE
(DRY SPECIMEN)

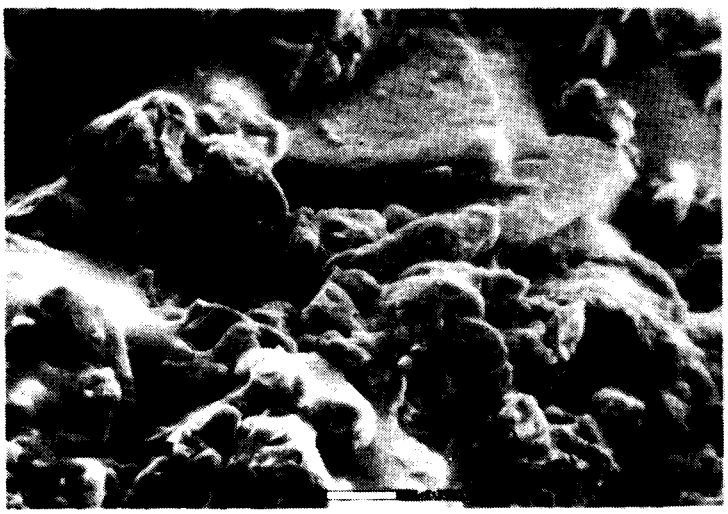

Fig . 9 - RES. 1 CORE SAMPLE
WATER SATURATED

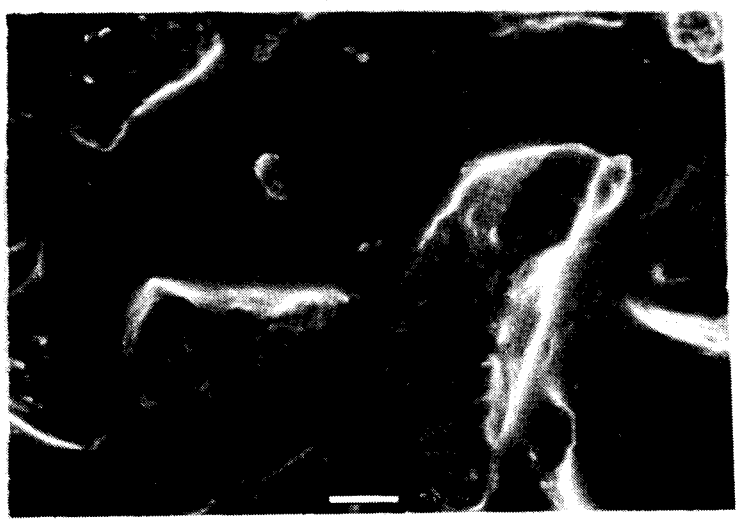

Fig .10 - RES. 2 CORE SAMPLE
WATER SATURATED

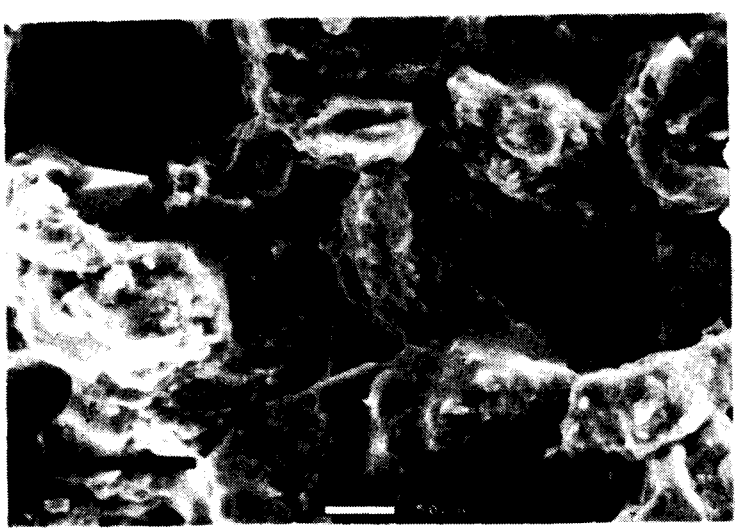

Fig .11 - RES. 3 CORE SAMPLE
WATER SATURATED

Fig .12 - FLUID DISTRIBUTION
AT Swi IN RES 1
CORE SAMPLE

Fig .13- FLUID DISTRIBUTION
AT Swi IN RES 2
CORE SAMPLE

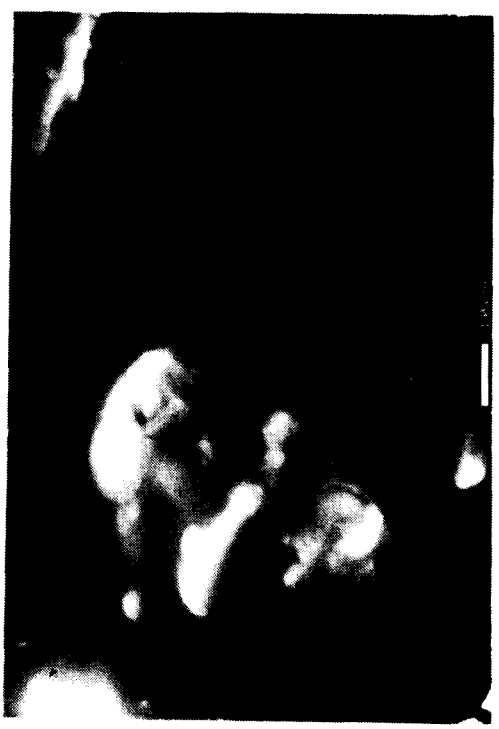

Fig .14 - FLUID DISTRIBUTION
AT Swi RES 3
CORE SAMPLE

Fig . 15 - FLUID DISTRIBUTION
AT Swi WATER
CONDENSATION IN RES 3
CORE SAMPLE

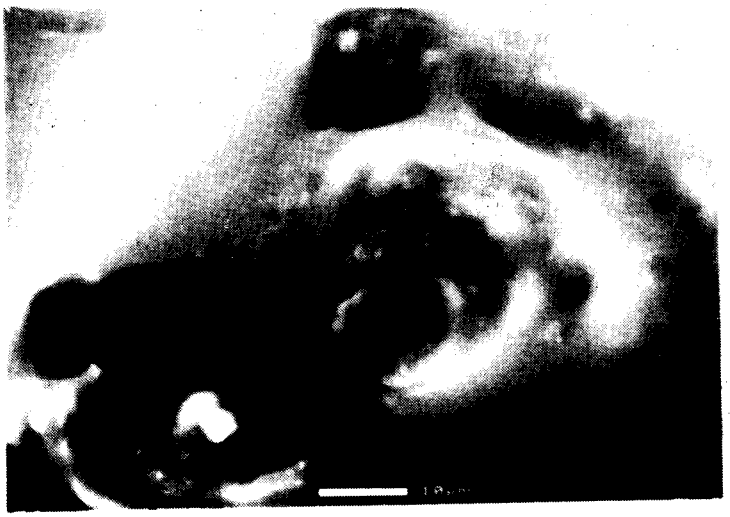

Fig . 16 - FLUID DISTRIBUTION
AT S_{OR} IN RES . 1
CORE SAMPLE

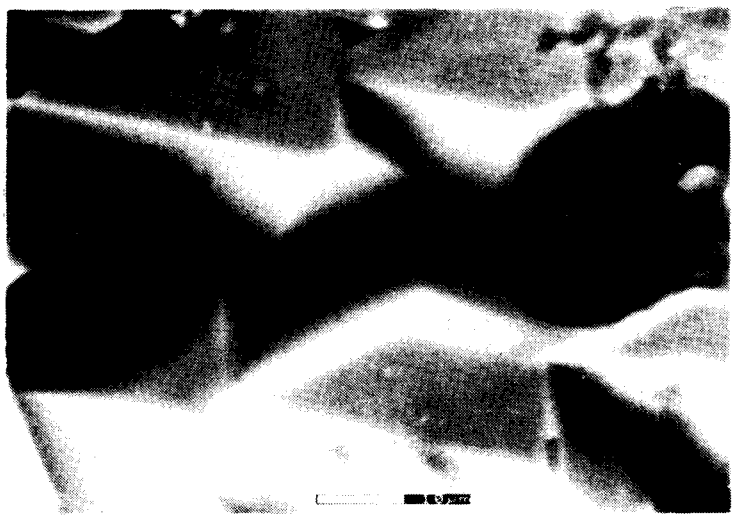

Fig . 17 - FLUID DISTRIBUTION
AT S_{OR} IN RES. 2
CORE SAMPLE

Fig .18 - FLUID DISTRIBUTION
AT S_{OR} IN RES . 3
CORE SAMPLE