ISSN: 1698-6180
www.ucm.es/info/estratig/journal.htm

Journal of Iberian Geology 32 (2) 2006: 197-213

Journal of
Iberian Geology
AN INTERNATIONAL PUBLICATION OF EARTH SCIENCES

Characterization of water pathways in low permeable rocks at the rock matrix scale: Methodological review

Caracterización de las vías de circulación del agua, a la escala de la matriz rocosa, en rocas de baja permeabilidad. Revisión metodológica

M. Montoto[1] and F. Mateos[2]*

[1] Dept. of Geology, Group of Petrophysics, University of Oviedo, 33005 Oviedo, Spain.
mmontoto@geol.uniovi.es
[2] GEA asesoría geológica. Uría 24- Oficina 9, 33003 Oviedo, Spain.
**Corresponding author: correo@geaasesoriageologica.com*

Received: 17/05/06 / Accepted: 03/08/06

Abstract

This is a methodological paper related to the characterization of water pathways, at the rock matrix or intact rock scale, in crystalline rocks candidate for the final disposal of high level radioactive wastes. At that scale, water circulation in these rocks is basically conditioned by its capacity to flow through the open effective porosity. That is, through the connected pores and fissures. Accordingly, the different procedures developed for imaging and mapping those pathways are here summarized. Among them, fluorescence microscopy, confocal laser scanning microscopy, scanning electron microscopy (secondary and backscattered electrons, and energy dispersive X-ray analysis), ^{14}C-polymethylmethacrylate impregnation,... stand out. In addition, the two main techniques for three-dimensional reconstruction of the pore space structure, X-ray computerized tomography and confocal laser scanning microscopy are also mentioned, in special the last one with different examples. The in-situ characterization of the rock porosity and the identification of paleo-water pathways, such as uranium induced fission track micro-mapping, are also described.

The main geometrical petrographic parameters related to the pore space structure are summarized and examples are also included, al□

Finally, examples of petrophysical profiles containing sequential information at the granulometric scale (mm-cm) are included; their objective being is to document in detail the capacity for water and radioelements to flow into the rock matrix from hydraulically active fractures in the rock mass.

Keywords: Water pathways, Paleo-water pathways, Intact rock, Rock matrix, Porosity, Pore space structure, Paleo-fissures, Petrophysical profiles, Rock imaging.

Resumen

Esta publicación metodológica está relacionada con la caracterización de las vías de circulación del agua, a escala de la matriz rocosa (o “intact rock”), en rocas cristalinas candidatas para almacenar definitivamente residuos radioactivos de alta actividad. A esta escala la circulación del agua en estas rocas está condicionada básicamente por su capacidad de fluir a través de la porosidad abierta efectiva. Es decir, a través de los poros y de las fisuras conectadas. Por consiguiente, se resumen los diversos procedimientos desarrollados para la obtención de imágenes y cartografía de estas vías de circulación, entre los que destacan: microscopía de fluorescencia, microscopía láser confocal, microscopía electrónica de barrido (electrones secundarios y retrodispersados, análisis por energía dispersiva de rayos X) e impregnación con ^{14}C-polimetilmetacrilato. Se mencionan las dos técnicas esenciales para la reconstrucción tridimensional de la estructura del espacio poroso, tomografía informatizada de rayos X y microscopía láser confocal y se incluyen diversos ejemplos de esta última. Además se describe la caracterización "in-situ" de la porosidad y la identificación de antiguas vías de circulación del agua mediante trazas de fisión del uranio.

Se resumen y se incluyen ejemplos de los principales parámetros petrográficos relacionados con la estructura geométrica del espacio poroso, ya que esta información es básica para la interpretación petrofísica de la función hidráulica de la porosidad de las rocas cristalinas.

Finalmente, se incluyen ejemplos de perfiles petrofísicos que contienen información secuencial a escala granulométrica (mm-cm), al objeto de documentar detalladamente la capacidad del agua y los radioelementos para introducirse y circular por la matriz rocosa a través de las fracturas hidráulicamente activas del macizo rocoso.

Palabras clave: Vías de circulación de agua, Paleo-vías de circulación de agua, Intact rock, Roca matriz, Matriz Rocosa, Porosidad, Estructura del sistema poroso, Paleo-fisuras, Perfiles petrofísicos.

1. Introduction

Finding a reliable solution to protect the biosphere from the radiotoxicity irradiated by high level radioactive wastes (HLW) is the main problem concerning those wastes. The internationally accepted solution for this problem is the isolation of these wastes in the interior of rock masss, under long-term requirements of stability and water tightness, until that radiotoxicity will be decayed to human acceptable doses.

That isolation is based on a "multibarrier concept" constituted by different engineered and natural systems; basically: canister, buffer and rock. In this multibarrier system, the selected rock mass plays one of the most important roles. In fact, for the very long-term basis, it is the final effective barrier for the wastes stored there.

As long as the principal medium for radioelement transport is water, hydrogeological studies are indispensable for the full characterization of the disposal site. According to all the studies realised up to now, a better understanding of the mechanisms governing radioelement migration in a geological scenario has been obtained. In any case, uncertainties still exist about the in-situ character of transport pathways at the intact rock scale. Therefore, more studies are required, particularly those related to the in-situ characterization of the pore space and the fluid flow (Frieg *et al.*, 1998).

As a consequence, in the mentioned framework of HLW studies, the water movements and their pathways through the rock mass deposit is one of the key questions to be understood and predicted. The objective of this publication follows that goal but focused on the intact rock scale. It tries to summarize the hydraulic function of the pore space and the procedures developed for the imaging and characterization of the water pathways, as well as their textural and mineralogical location. That is, their mapping. Therefore, this is a methodological paper related to this subject, not one providing specific results and conclusions concerning a specific geological site.

It is obvious that two basic scales of study coexist in any geological scenario: the rock mass scale (with fractures from a few m up to several km long) and the intact rock scale (with fissures and cracks to the order of µm to dm). The last one was defined by the "International Society for Rock Mechanics" referring to a rock volume free of massif discontinuities. The integration of data from both scales is a question that in Engineering Geology requires further development and many authors are aware of this problematic geological reality. The attempts for reproducing this situation in the laboratory, combining information from both scales, are very promising (Montoto, 2003). For instance, under this approach rock cores which include rock mass features such as fractures are studied; basically, non-standardized tests are being used for characterizing the hydraulic properties, and non-destructive procedures are applied for a preliminary characterization of rock anisotropy and heterogeneity.

At the intact rock scale, water circulation is basically conditioned by its capacity to flow through the connected pores and fissures (Fig. 1). In consequence, the different types of porosity have to be evaluated and hydraulically interpreted. The key aspects required for a deeper study and understanding of the hydrogeological behaviour of the rock-matrix and to characterize the textural and mineralogical location of water pathways, can be found in Montoto (2003). Among them, the following stand out: a) the two really different scales of porosity and water pathways coexisting in any geological scenario: the rock mass and the intact rock scale, b) fractographic parameters, such as fissure trace orientation, fissure volume, fissure range aperture, fissure network connectivity, fissure tortuosity, fractal dimension of fissure trace profiles,

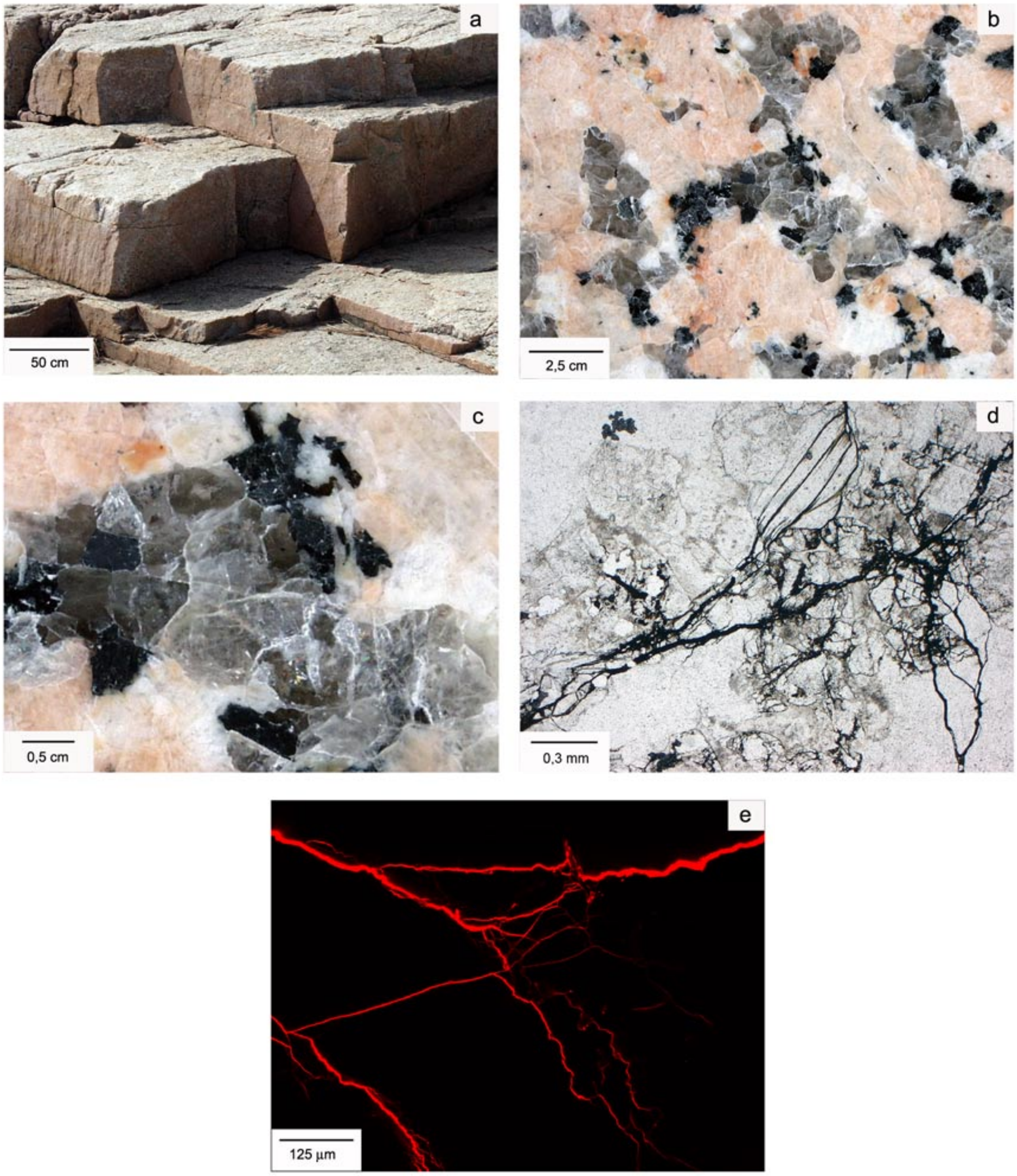

Fig. 1.- Fractographic features, water path ways, at the intact rock, or rock matrix, scale. (a) The rock mass discontinuities (fractures, joints,…) delimite blocks of intact rock of different size and shape; (b) close-up of the rock matrix observed in a flat polished slab; (c) detail of (b), intragranular cracks in quartz and feldspar are here visible at the nacked eye; (d) the well developed fractographic networks can also be identified under polarizing microscopy; and (e) the best procedure to observe the geometry, connectivity, tortuosity and flow potential of water path ways is laser confocal microscopy.

Fig. 1.- Características fractográficas, vías de circulación de agua, a la escala de la roca matriz o "intact rock". (a) Las discontinuidades de macizo (fracturas, diaclasas, ...) delimitan bloques de roca matriz de diferentes tamaños y formas; (b) aspecto macroscópico de la matriz rocosa observada en sección plana pulida; (c) detalle de (b), a simple vista pueden observarse fisuras intragranulares en cuarzo y feldespato; (d) las redes fractográficas mejor desarrolladas pueden observarse en microscopía de polarización; y (e) el mejor procedimiento para reconocer la geometría, conectividad, tortuosidad y capacidad de flujo de las vías de circulación del agua es la microscopía laser confocal.

and so on..., c) application of different microscopic techniques which provide images formed under very different "illumination radiations": photons, electrons, acoustic waves, electrical fields (polarized light microscopy, cathodoluminescence microscopy, fluorescence microscopy, confocal laser scanning microscopy, acoustic microscopy, scanning electron microscopy (secondary and backscattered electrons, and energy dispersive X-ray analysis), uranium induced fission track micro-mapping), d) quantitative microscopy procedures to map and evaluate the water pathways versus rock texture and mineralogy, as well as other rock-forming components, e) X-ray computerized tomography for the three-dimensional reconstruction of the pore space structure and f) preparation of artefacts-free rock thin sections. A very detailed description of all those techniques, images they provide, type of information they contain, applications to HLW, bibliographic references, etc. can be found in the afore mentioned reference.

Another basic interest for the characterization of the water pathways through the rock matrix and the quantification of their trajectories through the main rock-forming minerals is based on the potential fluid-solid reactions that can be developed in the rock; those reactions are responsible for radioelement retention by the rock-matrix, because a phenomenon developed on the interface between some rock-forming mineral surfaces and water conductive fissures. This process constitutes the basis for the positive results provided by the well known "rock matrix diffusion". Its importance is due to its particular role as a potential security factor in a repository. To illustrate the petrographic information of interest in this characterization, some examples about petrophysical profiles have been here included.

2. Water pathways through the intact rock

2.1. Hydraulic function of the connected pore space structure.

Water flow, at the rock-matrix scale is basically restricted through the open, connected, pore space structure and is conditioned by the geometry of the connections among pores and fissures and by the external or internal stresses affecting those channels. Similarly, at the larger rock mass scale, water circulation is mainly routed through the open rock mass discontinuities, such as faults, fractures, etc., which are generally referred as rock mass hydraulically active fractures.

To understand the hydraulic behaviour of the rock matrix, the most immediate required information refers to: a) total porosity (n), b) closed porosity (n_c) and c) open porosity (n_o); the last one being the most significant. It is used in Hydrogeology and subdivided into effective porosity (n_e) and trapped or non-effective porosity (n_t). The difference between "effective porosity" and "non-effective porosity" is instrumentally established at the very short term (in the order of some few days), according to the difficulties or practical impossibility of water movement through the rock. That difference is obviously of maximum interest. Unfortunately, does not exist any standard for this test; their values for a very short term response being deduced through Hg-porosimetry and then extrapolated to water behaviour. On the other hand, closed, or not connected, pores do not play any role in water flow. For instance, aligned closed pores in the interior of quartz grains can be found frequently, correspond to old fluid inclusions and, besides many other geochemical, chronological information, can inform about the paleo hydraulically active microfractographic network of the rock. That is about its paleo water pathways.

About the values of the open porosity in low permeable crystalline rocks, they use to be very low, in general less than 0,5 - 1%; that is, the lowest that can be found in rocks. The opposite happens in high permeable cemented rocks, with n_o values normally higher than 15 or 20%, and sometimes 30% or, exceptionally, even more.

As a practical conclusion, for a petrophysical understanding of the hydraulic function of the pore space of crystalline rocks, its geometry and volume has to be evaluated and the local direction and the values of geostresses, when possible. Consequently, some basic geometrical petrographic parameters should be obtained: a) fissure trace orientation; b) fissure volume; c) range of size of fissure apertures; d) percentage of the total effective porosity that at the very short term is refilled by water flowing through each aperture size; e) specific surface of the open fissure walls and f) specific surface of the open fissure walls in contact with each of the main rock-forming minerals (Montoto, 2003). Besides, fissure network connectivity, fissure tortuosity and fractal dimension of fissure trace profiles have to be considered in more specialized studies (Johns *et al.*, 1993; Pyralk-Nolte *et al.*, 1997; Montoto *et al.*, 1999a, b; Montoto, 2000a, b; etc.).

Knowing the variations in fissure aperture along their path orientation is a key aspect to understand potential local variations in permeability due to local geostresses. Analogously, due to the thermal stresses developed in the near field of a final repository, the evolution of the mentioned fissure aperture will affect water permeability. Therefore, both the pore space and the potential water mobility at the short- and long-term have to be predicted.

The knowledge of tortuosity and connectivity is also fundamental to understand water mobility (Fig. 2). Obviously, if water circulates through tortuous, narrow and poorly connected conduits, it would move slowly and with great difficulty at the short term scale. In addition, the effect of "ordered water layers" will contribute to make difficult the access of water and transported radioelements to the mineral walls of the void space. For that reason the expression "viscous" water is sometimes applied.

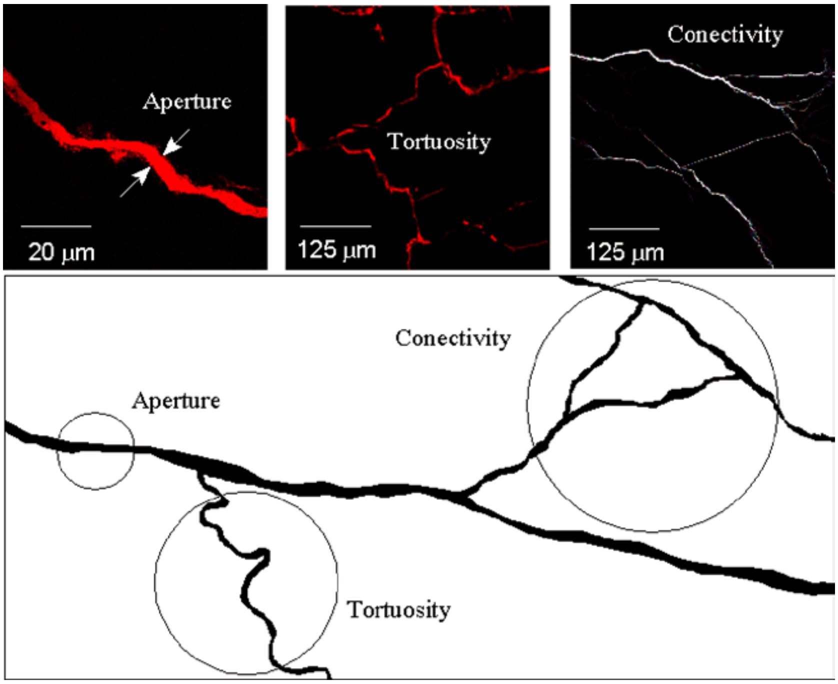

Fig. 2.- Sketch of the basic geometrical characteristics of a microfractographic network for water flow through the intact rock. The easiness or the difficulty for that flow is mainly controlled by the fractographic parameters: aperture, tortuosity and connectivity, as observed under confocal laser scanning microscopy.

Fig. 2.- Esquema de las características geométricas esenciales de una red microfractográfica para el flujo de agua a través de la matriz rocosa. La facilidad o dificultad para dicho flujo está fundamentalmente controlada por los parámetros fractográficos: anchura, tortuosidad y conectividad, tal y como se observa mediante microscopía láser confocal.

In addition to all the parameters up to now considered, the specific surface of the fissure walls in contact with each of the main rock-forming minerals is of a great significance in “rock matrix diffusion”, one of the most considered processes in natural analogue studies. This process, closely linked to water mobility through rocks, is conditioned by specific geochemical and petrophysical circumstances that could represent an added security factor played by the intact-rock. That is, in case of failure of the canisters and, also, in case of access of water to them and further lixiviation of radionuclides, the petrophysical and geochemical characteristics of the intact-rock could cause retardation in the transport of those radionuclides or, even their immobilisation. Consequently, for a better prediction of this type of long-term reactivity processes, to map the potential water pathways of the rock-matrix and to evaluate their spatial relationships to the different rock-forming minerals has been previously recommended (Montoto, 1996b, 2000a, b).

After all these considerations, it is clear that one of the most useful petrographic information of hydraulical significance is the variation range of the apertures of the water path ways. One of the most used instrumental procedures to evaluate the mentioned range of pore throat apertures is Hg-intrusion porosimetry, as well as gas adsorption. They are frequently complemented with quantitative petrographic microscopy techniques, such as stereology and digital image analysis. In the last case, for the three-dimensional reconstruction of the pore space structure they are used. Recently, X-ray computerized tomography is increasingly applied with the same objective (Ketcham and Carlson, 2001; Van Geet *et al.*, 2003; Vervoort *et al.*, 2004), as well as, autoradiography and polymethylmethacrylate impregnation (Siitari-Kauppi, 1995).

2.2. Present- and paleo-water-path ways.

The typical studies of the pore space structure of a rock obviously provide information about the present water pathways. The opposite have to be considered during the studies performed in Natural Analogues; their water pathways developed in the past have to be studied to understand the fluid-solid reactions occurring there. To evaluate the potential capacity of those reactions and their efficiency for radionuclide retardation and retention, the textural and spatial relationships between water pathways and rock-forming minerals have to be measured. Petrophysical and geochemical profiles, latter explained, also provide very useful information.

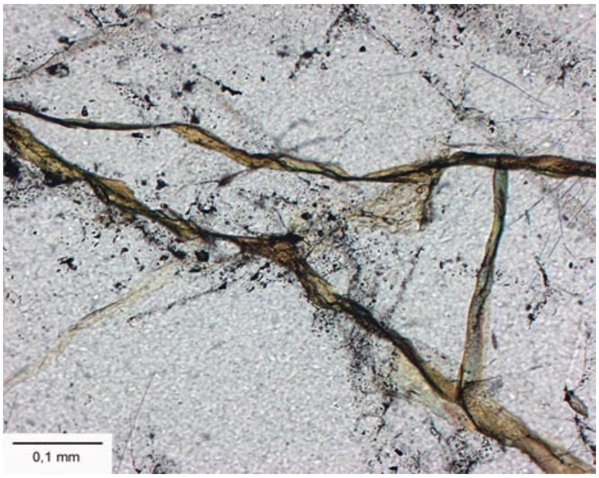

Fig. 3.- Paleo water pathways coated by Fe lixiviated from biotite in a weathered granite.

Fig. 3.- Paleo-vías de circulación de agua tapizadas por Fe procedente de la lixiviación de la biotita de un granito meteorizado.

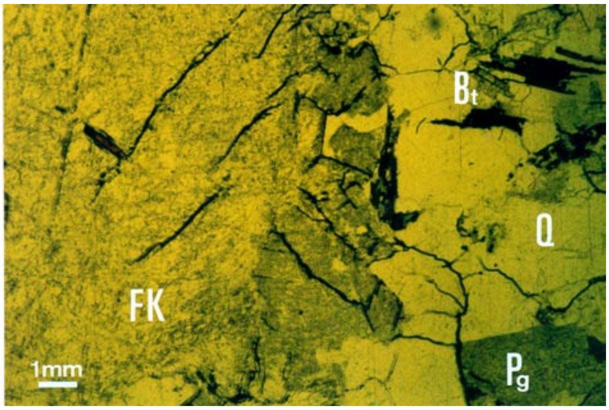

Fig. 4.- Paleo-fissures in granite mapped using U^{235} fission tracks, overlaying an image under polarizing microscopy. FK: K-feldspar, Bt: biotite, Q: quartz, Pg: plagioclase. El Berrocal (Toledo, Spain) granite. (Courtesy M. T. Ménager, Centre d'Etudes Nucléaires de Fontenay-aux-Roses, CEA, France).

Fig. 4.- Cartografía de paleo-fisuras en granito usando el método de trazas de fisión del ^{235}U superpuesta a una imagen de la misma zona bajo microscopía de polarizacion. FK: feldespato-K, Bt: biotita, Q: cuarzo, Pg: plagioclasa. Granito de El Berrocal (Toledo, España). (Cortesía de M. T. Ménager, Centre d'Etudes Nucléaires de Fontenay-aux-Roses, CEA, Francia).

The mentioned paleo-water pathways can be identified by the natural processes developed through them: oxidation mechanisms (Fig. 3), migration of radionuclides, etc (Ticknor *et al.*, 1986 a, b; Vandergraaf *et al.*, 1982; Kamineni *et al.*, 1986). Techniques for fissure decoration are described in Simmons and Richter (1976) and Pirhonen (1990).

The procedures for imaging fission tracks, (Menager *et al.*, 1982, 1994; Pérez del Villar *et al.*, 1994) allow a very precise mapping of U distribution in rocks at mm scale and the old paths followed during their migration (Fig. 4). The information they provide has been optimized using confocal laser scanning microscopy (Petford and Miller, 1990, 1992, 1993). Fission tracks are produced by neutron irradiation of ^{235}U after selective registration in an external detector which is tightly fixed to the polished rock thin sections. The distribution of the obtained tracks, uranium-enriched sites, can be related to the rock texture and mineralogy when compared with other microscopical images of the same area (Fig. 4). When the uranium appears as lineal shaped distribution located along fissures, cleavages, grain boundaries,... it has a special significance in paleo-microfractography hydraulically active, because it represents old migration pathways for water and radionuclides. (Perez del Villar *et al.*, 1995; 2000). In addition, any technique capable of mapping the geochemical elements present in rocks or, more precisely, in the interior of rock-forming minerals, is of prime interest for this objective; for example, electron microprobe, or the SEM+EDAX combination.

The example in figure 5 is a set of electron images which show a uraninite crystal, partially pseudomorphized by carbonates, and a long microfissure crosscutting the rock matrix from the source term to a small area where U-silicates have been precipitated. The microfissure is mainly filled by calcite, with U-silicates and idiomorphic pyrite. This microsystem in the rock matrix can simulate the source term (uraninite), the remobilisation agent (carbonate), the migration pathway (fissure), the migration mechanism (advective), the retention process (mainly by precipitation) and the transport distance. Furthermore, the physicochemical conditions under which the remobilation/migration/retention process took place can also be approximately inferred (Pérez del Villar *et al.*, 2000).

2.3. In situ characterization of the intact rock porosity

The microfractographic analysis and the evaluation of rock porosity and water pathways has been classically and routinely performed on laboratory experiments (Kowallis and Wang, 1983; Hellmuth *et al.*, 1995; Rasilainen *et al.*, 1996; Mazurek, 2000). In these experiments, specimens from rock cores drilled at very different depths have been used. As it has been extensively proved, those specimens contain fractographic artefacts developed during drilling, recovery of cores from deep boreholes, preparation of specimen and rock thin sections, etc. Under those circumstances, a clear uncertainty can be argued about how the obtained results inform about the *in situ* reality of the rock matrix pore structure. In consequence, a serious question remains: are those results acceptable to interpret the in-situ hydraulic properties of the intact rock? (Mazurek, 2000).

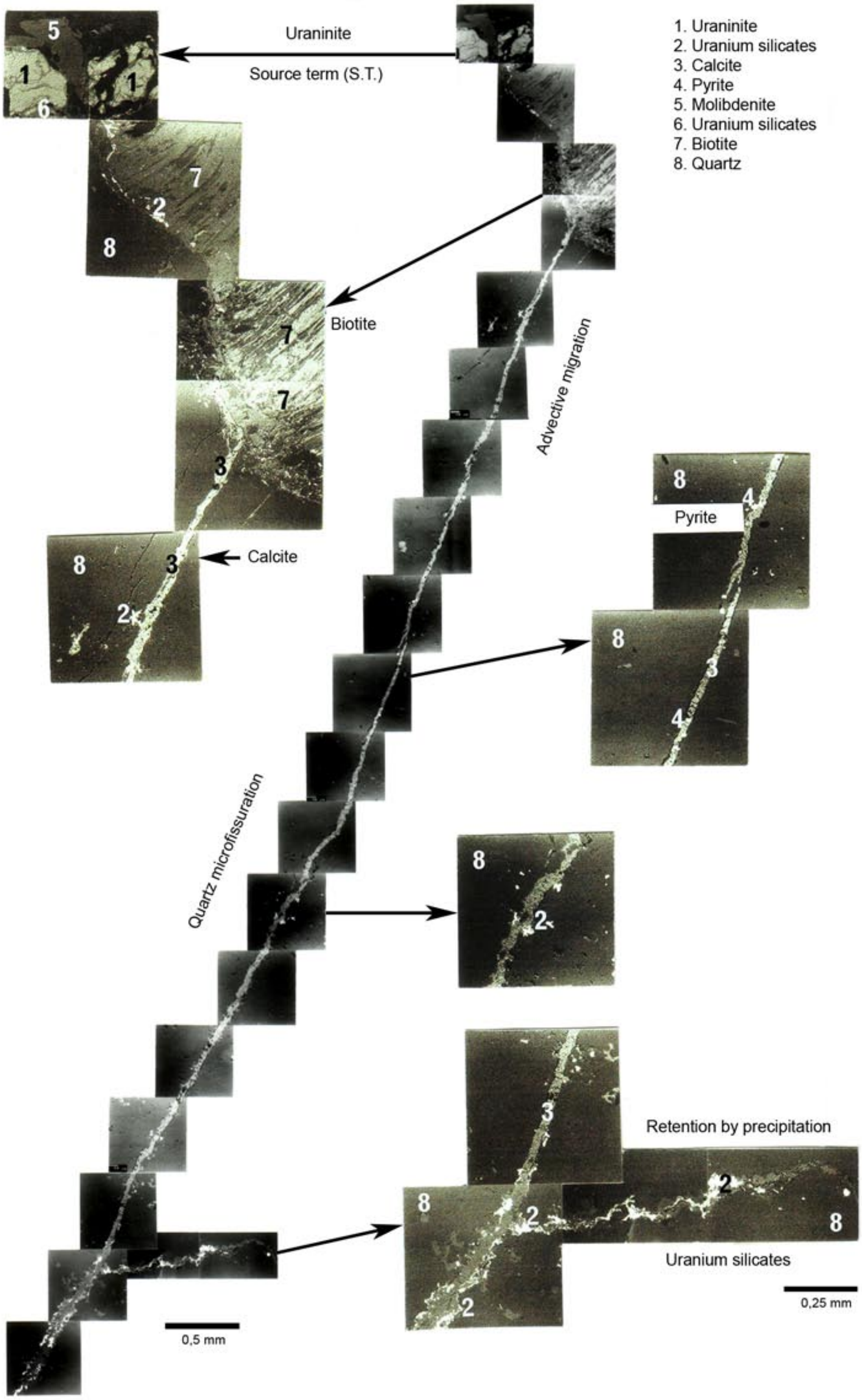

Fig. 5.- Set of backscattered electron images showing a microfissure, mainly intersecting a quartz grain which joins the source term (uraninite 1) with a small area where U silicates (2) have precipitated. The microfissure is mainly filled by calcite (3), with U silicates (2) and idiomorphic pyrite (4). Molybdenite (5), U-carbonates ? (6), weathered biotite (7) and quartz (8). Sample from the U-rich granite of Palmottu. Courtesy of L. Pérez del Villar and J. S. Cózar (CIEMAT, Centro Investigaciones Energéticas y Medio Ambientales; Madrid, Spain).

Fig. 5.- Conjunto de imágenes electrónicas (electrones retrodispersados) mostrando una microfisura que intersecta principalmente a un grano de cuarzo y que comunica el término fuente (uraninita 1), con una pequeña área donde han precipitado los silicatos de U (2). La microfisura aparece principalmente rellena de calcita (3), con silicatos de uranio (2) y pirita idiomórfica (4). Molibdenita (5), carbonatos de U? (6), biotita alterada (7) y cuarzo (8). Muestra procedente del granito uranífero de Palmottu (Finlandia). Cortesía de L. Pérez del Villar y J. S. Cózar (CIEMAT, Centro Investigaciones Energéticas y Medio Ambientales; Madrid, Spain).

To avoid the existing uncertainties in transport pathway, and for a more realistic modelling of rock matrix diffusion process, the in-situ characterization of the connected porosity has been promoted by NAGRA (National Cooperative for the Disposal of Radioactive Waste, Switzerland) and PNC (Power Reactor and Nuclear Fuel Development, Japan) (Frieg *et al.*, 1998). That research has been programmed in the vicinity of a water conducting fracture in the Nagra's underground rock laboratory (Grimsel Test Site, GTS), using resin impregnation techniques.

It is clear that the mentioned *in situ* impregnation represents a great advance on the characterization of the original rock matrix porosity and pore space geometry (Schild *et al.*, 2001). These authors studied the GTS 3-D fractographic network using microscopical and ultrasonic techniques. In-situ fluoresceine impregnated rock thin sections were thus analyzed under fluorescence microscopy equipped with a Universal stage; the existing rock cracks were classified according to their host grain, textural position (inter or intragranular) and state (open, healed, and sealed), (Schild, 1999). Finally, they were evaluated in orientation and frequency. Furthermore, the *in situ* cracks were discriminated from those formed during core relaxation and sample preparation. The final and very important conclusion is that the previous data on porosity of the Grimsel granodiorite conventionally measured in the laboratory 0.8 - 1.2% (Bossart and Mazurek, 1991) has to be decreased to about 0.4 - 0.5%, which represents a significant difference.

3. Imaging the water pathways

To contribute to a better understanding of migration of radionuclides through rocks at any scale, it is fundamental to observe their pathways and their textural and mineralogical situation. This approach allows a better understanding and evaluation of the specific role played by each rock forming component in retention processes such as "rock matrix diffusion".

The imaging, mapping and quantification of the open effective porosity in rocks, have been classically performed under fluorescence microscopy (FM) and the different techniques mentioned in the Introduction. From the 90's new techniques and procedures: confocal laser scanning microscopy (CLSM), ^{14}C PMMA impregnation technique and microfocus X-ray computer tomography (µCT) have been developed for those objectives; and more specifically, for a 3D imaging of the water pathways that, at the intact rock scale affects the candidate crystalline rocks for the final disposal of HLW.

The ^{14}C-PMMA method involves the impregnation of the rock with this labelled organic compound in a vacuum, irradiation polymerisation, autoradiography and optical densitometry, using digital image-processing techniques (Hellmuth *et al.*, 1993, 1995, 1999, Siitari-Kauppi, 1995 and Siitari-Kauppi *et al.*, 1998a, b). It provides information at the nm – cm scale about the accessible pore space in low permeable crystalline rock that cannot be obtained using other methods. MMA molecule intrudes into the nanometric scale pores, and the autoradiography allows the visualisation at 10 µm resolution.

Microfocus X-ray computer microtomography (X-ray µCT) is another extremely promising technique for imaging the rock-forming components in the interior of the rock matrix. As a strong contrast exists between the rock-forming minerals and the voids in the images of a rock under X-ray µCT, the study of the pore space can be performed using this technique (Perret *et al.*, 1999; Van Geet *et al.*, 2003; Karakan *et al.*, 2003; Vervoort *et al.*, 2004). In addition, it can provide information of the movement of water through the rock matrix (Anderson *et al.*, 2003; Géraud *et al.*, 2003; Ruiz de Argandoña *et al.*, 2003; Sugawara *et al.*, 2004). It can also reveal details of the 3D structure of the water pathways, and of their textural and mineralogical position, etc. Therefore the technique is also capable of provide qualitative and quantitative analysis of internal features of geological materials, if those features are marked by sufficiently great differences in atomic composition and/or density (Mees *et al.*, 2003). The results provided by this technique are very often complemented with others obtained under optical microscopy and scanning electron microscopy (Van Geet *et al.*, 2001).

Probably, fluorescence microscopy (FM) is the most routinely used technique among those previously mentioned; not only for tradition and its lower instrumental costs, but also for its advantage of using the same rock thin sections for further textural and mineralogical studies. In fact, the combination of polarizing microscopy and FM (Fig. 6) is the most recommended procedure for a petrographic mapping of open fissures in rocks (Montoto *et al.*, 1980).

Fluorescence microscopy (FM) requires a previous rock impregnation with a fluorescent dye penetrant and later the preparation of flat rock thin sections or slabs without disturbing its original characteristics. That is, avoiding the development of artefacts, such as new cracks. This basic requirement implies the use of specific procedures, such as low deformable sawing machines, grinding, etc. Then, those sections can be observed under FM, CLSM and SEM using secondary or backscattered electrons.

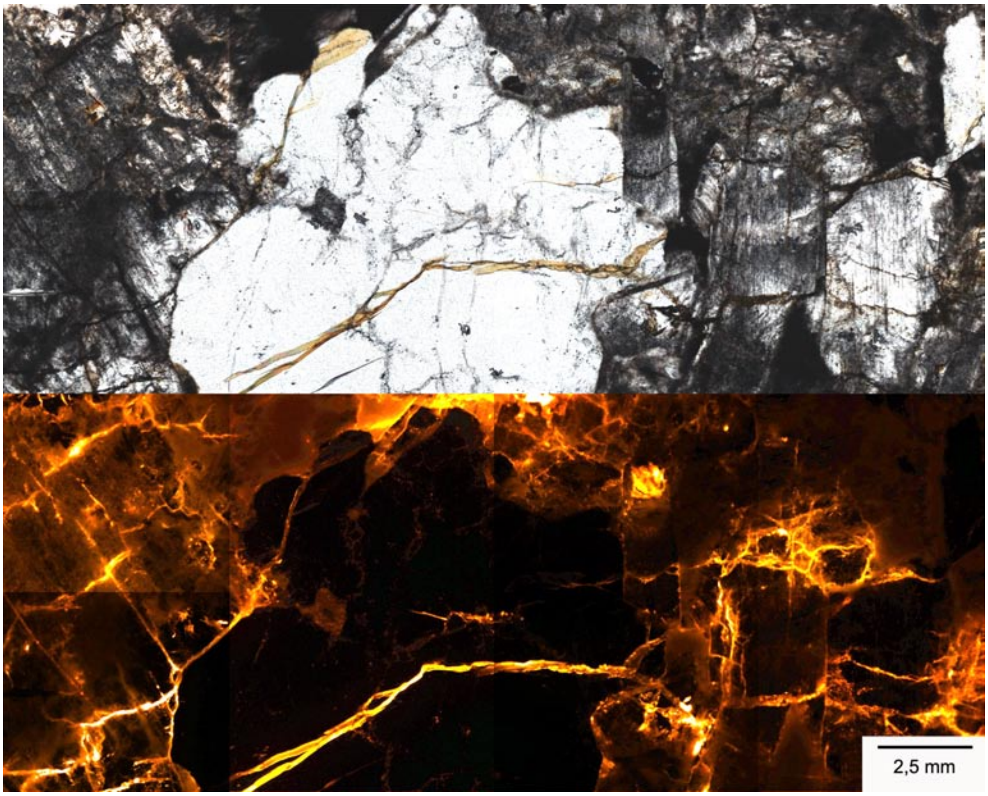

Fig. 6.- Mineralogical and textural mapping of water pathways in rocks under fluorescence microscopy and polarizing microscopy.

Fig. 6.- Cartografía mineralógica y textural de las vías de circulación del agua en rocas utilizando microscopías de fluorescencia y de polarización.

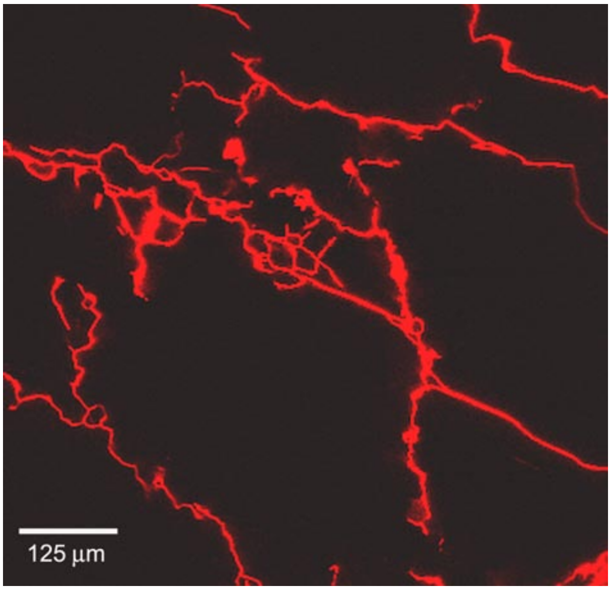

Fig. 7.- CLSM allows the observation of the geometrical complexity of the water pathways in crystalline rocks. Connectivity or tortuosity can be clearly identified and evaluated.

Fig. 7.- Mediante CLSM se evidencia la complejidad geométrica de las vías de circulación del agua en rocas cristalinas. La conectividad y tortuosidad pueden identificarse y cuantificarse.

This last technique can be also used with not impregnated rock slabs or thin sections, as well as fracture surface specimens. These specimens provide very interesting images but less helpful for mapping and quantification.

All the images obtained under the up to now mentioned microscopical techniques can be processed by means of digital image analysis procedures; the objective being a better discrimination of the impregnated open pore space and the grain mineral borders for an easier mapping of texture, mineralogy and water pathways.

In crystalline rocks, water circulation is restricted to fissured paths of intergranular, transgranular or intragranular character. This textural and mineralogical location of the water pathways has to be evaluated in terms of their specific surface in relation to the main rock-forming minerals.

CLSM is, undoubtedly, the best procedure to observe, at the µm – mm scale, the finest geometrical details (Fig. 7) of the open microfractographic network -rock water pathways- in crystalline rocks (Montoto *et al.*, 1995, Montoto, 1999e, f). If compared to FM, the CLSM results are outstanding, due to the higher lateral resolution of this technique. Besides, the three-dimensional observation of that geometry under CLSM provides information that nowadays can be considered as unique.

Sequential virtual sections –optical sections- can be obtained under CLSM, at regular distances of about 1 µm (Fig. 8). The complexity of the rock microfractographic network, in terms of connectivity and tortuosity, can be clearly identified. In general a three-dimensional reconstruction of those types of networks can be obtained by

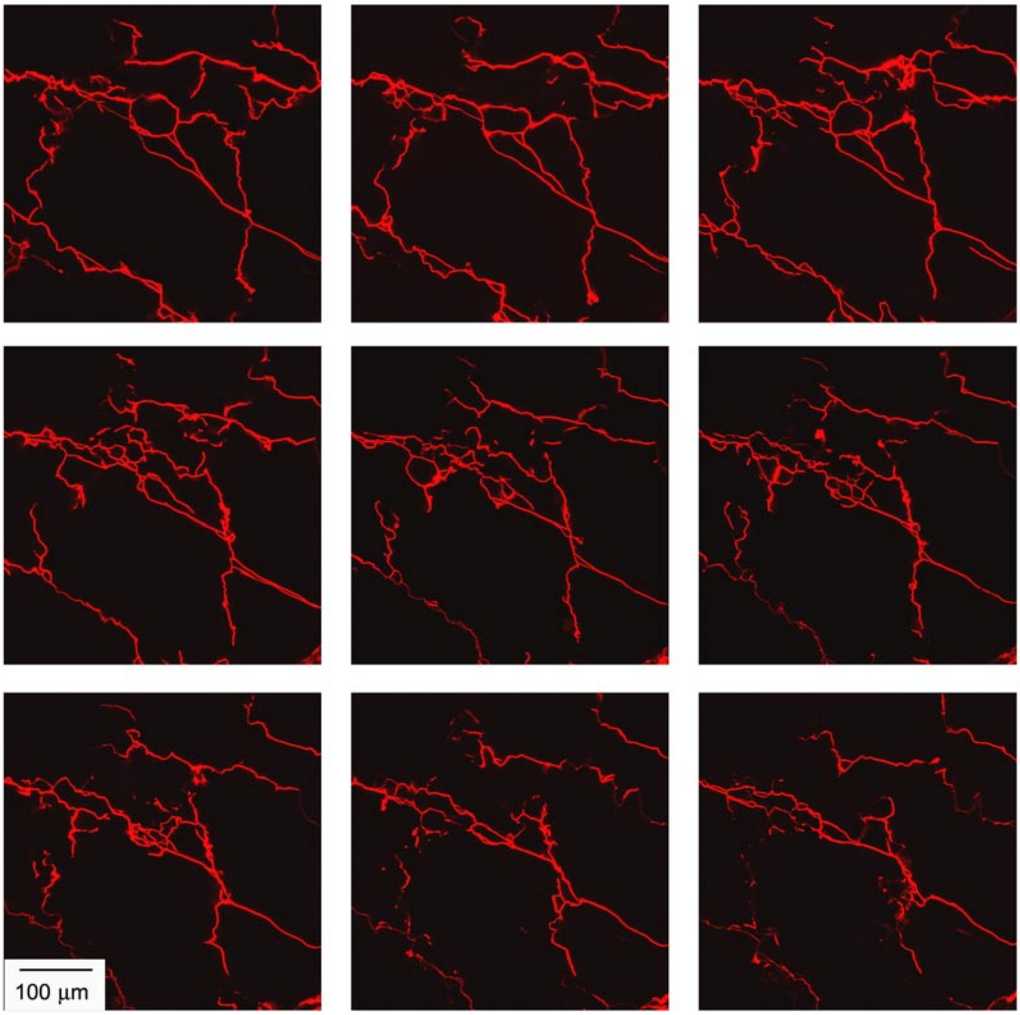

Fig. 8.- The geometric characteristics of the 3D network of water pathways in granitic rocks can be identified by means of sequential optical sections obtained under CLSM. In this example the 9 sections are 5µm apart. Data about fissure orientation, dip, connections, tortuosity, etc. can be automatically obtained.

Fig. 8.- Mediante CLSM pueden obtenerse secciones ópticas secuenciales paralelas entre sí e identificar con todo detalle las características geométricas de la red tridimensional de las vías de circulación del agua. En este ejemplo se muestran 9 secciones separadas entre sí 5µm. Los datos sobre la orientación, el buzamiento, las conexiones, la tortuosidad, etc. de las fisuras pueden obtenerse mediante procesos automatizados.

an appropriate processing of a large number of sequential optical sections (Fig. 9).

After the corresponding processing and 3D reconstruction of optical sections sequentially obtained under CLSM, an extremely precise visualization of the three-dimensional network of the water path ways can be obtained, as examples from the El Berrocal (Spain) site proves (Montoto, 2003). Each optical section is displayed in a different colour according to its depth in the rock section specimen and to an arbitrary colour-table code. Later those coloured images are superimposed and a new image is formed. In this image, a reading of the exact position, dip, orientation, connection, etc. of any water pathway, or open fissure, can be carried out. The whole three-dimensional reading of a given zone in terms of direction, dip, tortuosity and connectivity of the main water pathways can be easily performed. In addition, all the structural and microfractographic information, at the µm - mm scale, thus obtained can be related to any other geologic information of the rock-mass.

Specific examples of microfractographic studies under CLSM and ^{14}C PMMA related to the disposal of HLW can be found in Montoto *et al.* (1995, 1999c, d, f) and Fernández-Merayo *et al.* (1996). This technique offers the ability to collect serial optical sections (or virtual sections) from thick specimens by the elimination of image out-of-focus information. That is, the big advantage of confocal microscopy is the possibility to collect light exclusively

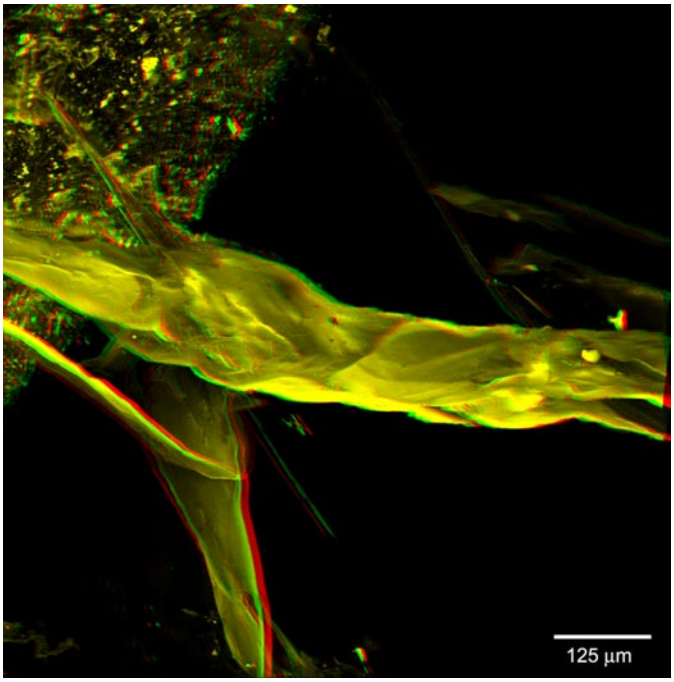

Fig. 9.- 3D visualization of the microfractographic network in a granite sample 80 optical sections have been sequentially obtained under CLSM, 2µm apart, and later processed and superimposed.

Fig. 9.- Visualización en 3D de la red microfractográfica en una muestra granito. Se han obtenido 80 secciones ópticas bajo CLSM separadas entre sí 2µm, que han sido procesadas y superimpuestas.

from a single plane of the interior of the rock thin section. In the procedure, a pinhole sitting conjugated to the focal plane (i.e. confocal) keeps light from the detector that is reflected /emitted from others than the focal plane. The CLSM scans the sample sequentially point by point and line by line and assembles the pixel information to one image; that way optical slices of the specimen are imaged with high contrast and high resolution in x, y and z. By moving the focus plane single images (optical sections) can be put together to build up a three dimensional stack (Fig. 10).

The combination of some of the mentioned techniques provides the optimum results. In a recent paper, Kelokaski *et al.* (2006) demonstrate the main advantages that two new techniques, CLSM and ^{14}C PMMA, offer for a better imaging and mapping of the water pathways, water bearing fissures, at the µm-cm scale, in low permeable crystalline (fissured) rocks. In addition, by complementing those two techniques a wide range of scales can be covered. The results of ^{14}C PMMA at the cm-dm scale are impressive and the same with CLSM at the µm-cm.

More information about water pathways and techniques for their identification can be found in: Menager *et al.*, (1982, 1994); Vandergraaf *et al.*, (1982); Ticknor *et al.*, (1986 a,b); Yoshida *et al.*, (1993, 1994); Degueldre *et al.*, (1996) and Johansson *et al.*, (1998).

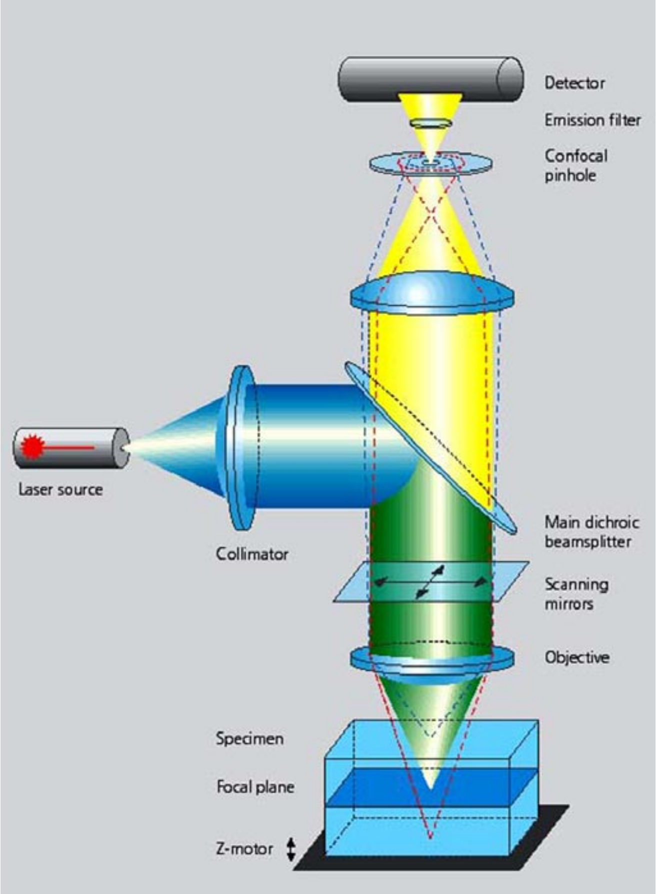

Fig. 10.- Sketch of the confocal laser scanning microscope and the procedure, confocal pinhole, to eliminate out-of-focus information. (The Confocal Laser Scanning Microscope, 2005). Carl Zeiss, www.zeiss.de.

Fig 10.- Esquema del microscopio láser confocal de rastreo y procedimiento para la eliminación de la información “no enfocada”, mediante el diafragma “confocal”. (The Confocal Laser Scanning Microscope, 2005). Carl Zeiss, www.zeiss.de.

4. Petrophysical profiles

As it has been previously mentioned, two really different scales of porosity and water pathways coexist in a geological scenario: those scales correspond to the rock-mass and to the intact rock; for this reason, the expression double-porosity medium is so precise and representative in hydrogeological studies. In relation to those circumstances, a very interesting aspect of application in the disposal of HLW is the role played by the intact rock in its immediate contact with a water bearing fracture of the rock mass.

If the most adverse conditions in a HLW repository are imagined and the established multi-barrier system fails, the radioelements could be mobilised from the canisters and transported trough the rock-mass. Fortunately, under given hydrogeochemical conditions those radioelements

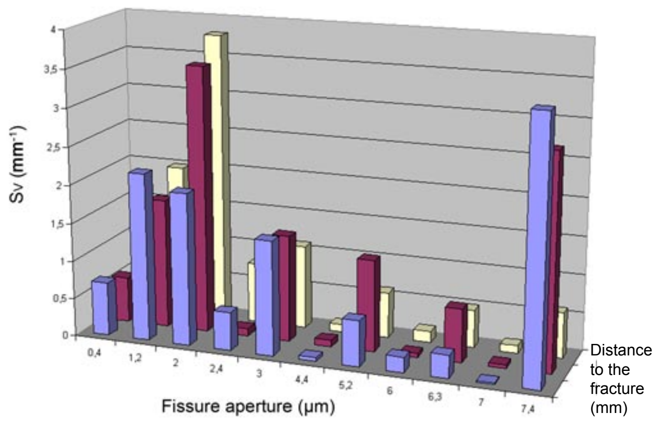

Fig. 11.- Profile of the geometric characteristics of the water pathways in the intact rock in its immediate vicinity of a water bearing rock-mass fracture: a) specific surface of open fissure walls, b) aperture of open fissures and c) distance to the fracture. El Berrocal granite, Toledo, Spain. (Montoto *et al.*, 1992).

Fig. 11.- Perfil de las características geométricas de las vías de circulación del agua en la matriz rocosa, en la proximidad inmediata a una fractura hidráulicamente activa del macizo. a) superficie específica de las paredes de las fisuras abiertas, b) anchura de las fisuras abiertas y c) distancia a la fractura. Granito de El Berrocal, Toledo, España. (Montoto *et al.*, 1992).

could be retarded in their velocity of transport or, even, immobilised into the rock matrix. This radioelement retention capacity, usually termed rock matrix diffusion or rock matrix retardation, (Neretnieks, 1980; Hadermann and Rösel, 1985; Alexander *et al.*, 1990 a,b; Ivanovich and Harmon, 1992; Ivanovich *et al.*, 1994, 1987a,b) has been proven to be an added safety factor in the repository. That is, it contributes to the isolation of radionuclides within the rock mass, preventing them from reaching the biosphere.

This matrix diffusion phenomenon "flows" from rock-mass hydrogeologically active fractures towards the interior of the intact rock. Studies in natural analogues (Miller *et al.*, 1994, 2000; Gauthier-Lafaye *et al.*, 1997) prove that the mentioned retardation has been observed within the rock matrix, at distances of about 10-12 cm apart from those rock-mass fractures. Therefore, a great rock-mass volume contributes to this process and, in consequence, to evaluate its real dimension, many different type of data has to be obtained: hydraulic, physico-chemical, petrographic,… Among them, the capacity for water and radioelements to flow into the rock matrix from hydraulically active fractures, and the corresponding water pathways, is one of the main aspects to be considered. In addition, most of the features of the pore space structure, up to now mentioned for its hydraulical significance, have to be sequentially evaluated from the fracture surface to the rock interior. This sequential information constitutes the "profiles". Examples of petrophysical and geochemical profiles at the cm scale can be found in Heath *et al.* (1992); Montoto *et al.* (1996b, c); Montoto (1999a, e, f).

The potential solid-fluid reactivity, a key question in rock matrix diffusion, should be predicted. Therefore, besides other physico-chemical information, the spatial relationship existing between the water pathways and each of the rock-forming minerals has to be evaluated. For this reason, the water pathways are mapped and from those images, their specific surface related to each rock-forming mineral is evaluated using image analysis procedures. As explained before, all the mentioned information has to be presented in sequential order as "profiles.

Those profiles are obtained after a systematic data acquisition at established aligned distances from the wall of the fracture towards the interior of the intact rock. Profiles of very different content are thus obtained. For the objectives of this paper, the profiles particularly relevant are those concerning the water transference between that fracture and the internal pathways of the intact rock. Consequently, the most significant petrophysical data are: rock heterogeneity, rock anisotropy, local geostresses, water absorption capacity, capillarity coefficient at the very short term, open porosity, range and distribution of the apertures of open fissures, specific surface of the walls of open fissures, specific surface of water path ways related to each of the main rock-forming minerals, etc.

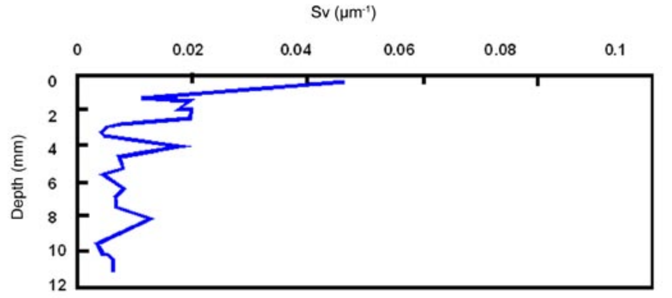

Fig. 12.- Profile of the Open Crack Specific Surface (OC S_v) evaluated using Stereology.

Fig. 12.- Perfil de la superficie específica de la totalidad de las fisuras abiertas (OC S_v), evaluada mediante estereología.

Equivalent studies with a very different purpose are usually performed in the excavated disturbed zones (EDZ). To understand the evolution suffered by the pore space structure and the new water flow capability through the intact rock is the main objective here. In the full-scale experimental deposition holes of the TVO research tunnel (Olkiluoto, Finland) site, many different techniques have been applied to analyze the effective porosity of the bored disturbed zone for the final disposal of canisters. Those techniques include: ^{14}C PMMA and He-gas (Autio *et al.*,

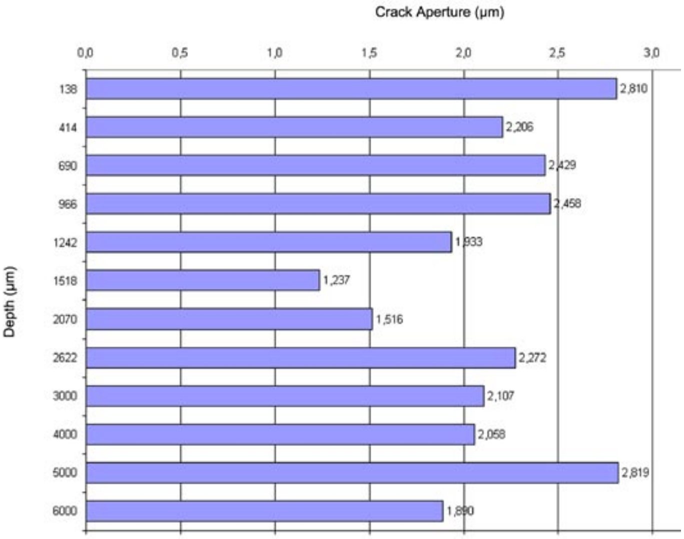

Fig. 13.- Profile of the Open Crack Volume percentage (OC V %) obtained under digital image analysis.

Fig. 13.- Perfil del porcentaje del volumen de la fisuración abierta, obtenido mediante procesado digital de imágenes.

1998) quantitative microscopy -stereology and digital image analysis (Montoto *et al.*, 1999c, d); etc. All the results obtained have been compared (Campos and Rivas, 2002) and the extent of the damaged zone has been found congruent. Rock-forming minerals and the new open fissures developed in the rock, potential water pathways, have been imaged under different microscopical techniques: polarizing microscopy, reflected light FM, CLSM, SEM and BSEM. The information contained in the images thus obtained has been quantified, using both stereology and digital image analysis techniques described in detail in Menéndez (1992) and Martínez-Nistal (1993).

Some examples of different petrophysical profiles at the mm-cm scale, related to geometric characteristics of the water path ways in applications to rock matrix diffusion and EDZ are presented in figure 11, 12 and 13.

The open crack specific surface (OC S_v) is the area of the walls of all the open cracks/fissures in the rock by volume (µm^2/µm^3); many times it is evaluated according to the textural position (inter, intra, etc.) of cracks in each of the main rock forming minerals. In granite, for example, this type of data is presented as: OC S_v (Q), OC S_v (F), OC S_v (M), referring to quartz, feldspar and mica, respectively A profile of the OC S_v in the full-scale experimental deposition holes of the TVO research tunnel Olkiluoto tonalite, Finland, (Montoto *et al.*, 1999b, c, d) is presented in figure 12. The values of the OC S_v from the border of the bored disturbed zone to the interior of the intact rock are plotted, and the new water flow capability, related to the gradual modification suffered by the pore space structure, can be analyzed.

The open crack volume percentage (OC V %) refers to the volume occupied by the open cracks/fissures. This parameter, in crystalline rocks, not in cemented rocks, is very often assimilated to the effective porosity of the intact rock. It is evaluated through the data of the crack/fissure apertures. In the figure 13, a profile of this parameter versus the distance to the border of the bored disturbed zone in the Olkiluoto tonalite is plotted.

5. Concluding remarks

As the principal medium for radioelement transport is water, hydrogeological studies are of prime interest to the full characterization of the disposal site of HWL. At the intact rock scale, the studies related to the capacity of water to flow through the connected pores and fissures contribute to that characterization. In addition, the identification of water pathways as well as their textural and mineralogical location is also very significant information. This mapping of water pathways is basic for a more completed petrophysical interpretation of the hydraulical behaviour of the intact rock. Furthermore, it contributes to a better prediction of “rock matrix diffusion” process, that is, the potential fluid-solid reactions responsible of radioelement retention by the rock-matrix, which in fact is a potential security factor in a repository.

For those reasons, the procedures for imaging and mapping water pathways and the appropriate parameters for their quantification are subjected to continuous improvement.

Fluorescent microscopy is the most classical technique for imaging water pathways, due to its higher lateral resolution. Nowadays, confocal laser scanning microscopy is the best procedure to observe, at µm-mm scale, the finest geometrical details of the open microfractographic network of a rock. The examples here included prove that CLSM provides unique information for a three-dimensional visualization of that network. For a really precise textural and mineralogical mapping of water pathways, CLSM can be combined with polarizing microscopy or scanning electron microscopy. Other modern techniques, such as ^{14}C-polymethylmethacrylate (^{14}C PMMA) impregnation technique and microfocus X-ray computer tomography (µCT), also provide extremely valuable information.

Among the different parameters appropriate to characterize the water pathways in crystalline rocks, the following stand out: a) fissure trace orientation; b) fissure volume; c) range of size of fissure apertures; d) percentage of total effective porosity that at the very short term is refilled by water flowing through each aperture size; e) specific surface of the open fissure walls and f) specific surface of the open fissure walls in contact with each of the main rock-forming minerals. Other data, such as fissure network connectivity, fissure tortuosity and fractal dimension of fissure trace profiles are also very helpful for a better understanding of water kinetics through the intact rock.

Finally, the capacity of water to flow from water bearing fractures of the rock mass into the rock matrix is a key aspect to be documented for a better understanding of the full hydraulical behaviour of any geological scenario. Therefore, sequential information or petrophysical profiles, at mm-cm scale, from those fractures towards the immediate interior of the rock matrix has to be provided. That information mainly refers to the hydraulic properties of the intact rock and the geometry of its water pathways.

Acknowledgements

To the Commission of the European Communities (Research and training in nuclear energy) and ENRESA (Empresa Nacional de Residuos S.A.); their co-financing of the FUNMIG Integrated Project (Fundamental processes of radionuclide migration; FI6W, 516514 and 0078000068) allows some of the research tasks on progress here summarized.

To Dr. L. Pérez del Villar (CIEMAT; Madrid, Spain) for his kind information, continuous help and corresponding example shown in figure 5.

References

Alexander, W.R., Mackenzie, A.B., Scott, R.D., McKinley, I.G. (1990a): *Natural analogue studies in crystalline rock: The influence of water-bearing fractures on radionuclide immobilisation in a granitic rock repository*. Nagra Technical Report 87-08: 42 p.

Alexander, W.R., McKinley, I.G., MacKenzie, A.B., Scott, R.D. (1990b): Verification of matrix diffusion by means of natural decay series disequilibrium in a profile across a water conducting fracture in granitic rock. *Scientific Basis for Nuclear Waste Management*, 23: 567-576.

Anderson, S.H., Wang, H., Peyton, R.L., Gantzer, C.T. (2003): Estimation of porosity and hydraulic conductivity from X-ray CT-measured solute breakthrough. In: Mees, F., Swennen, R., Van Geet, M., Jacobs, P. (eds.): *Applications of X-ray computed tomography in the geosciences.* Geological Society, London, Special publications 215: 135-149.

Autio, J., Siitari-Kauppi, M., Timonen, J., Hartikainen, K., Hartikainen, J. (1998): Determination of the porosity, permeability and diffusivity of rock in the excavation-disturbed zone around full-scale deposition holes using the ^{14}C-PMMA and He-gas methods. *Journal of Contaminant Hydrology,* 35: 19-29.

Gauthier-Lafaye, F., Blanc, P-L., Bruno, J., Griffauld, L., Ledoux, E., Louvat, D., Michaud, V., Montoto, M., Oversby, V., Pérez del Villar, L., Smellie, J. (1997): The last nuclear fission reactor. *Nature*, 387: 337.

Bossart, P., Mazurek, M. (1991): *Grimsel Test Site: Structural geology and water flow-paths in the Migration shear-zone*. Nagra Technical Report NTB 91-12, Nagra, Switzerland: 55 p.

Campos, R., Rivas, P. (2002): *Metodología aplicada al estudio de la variación de la porosidad y microestructura en la EDZ desarrollada en la Galería FEBEX (Grimsel-Suiza)*. CIEMAT-Departamento de Impacto Ambiental de la Energía. 50p. Proyecto HIDROFEBEX, CIEMAT/DIAE/54421/5/02.

Degueldre, C., Pleinert, H., Maguire, P., Lehman, E., Missimer, J., Hammer, J., Leenders, K., Böck, H., Townsend, D. (1996): Porosity and pathway determination in crystalline rock by positron emission tomography and neutron radiography. *Earth and Planetary Science Letters*, 104: 213-225.

Fernández-Merayo, N., Siitari-Kauppi, M., Montoto, M., Hellmuth, K-H. (1996): Characterization of rock matrix structure using microscopy ^{14}C-PMMA method for the study of retardation mechanisms in granite. *Radiochimica Acta,* 74: 211-215

Frieg, B., Alexander, W.R., Dollinger, H., Buhler, C., Haag, P., Mori, A., Ota, K. (1998): In situ resin impregnation for investigating radionuclide retardation in fractured repository host rocks. *Journal of Contaminant Hydrology*, 35: 115-130.

Géraud Y., Surma F. and Mazzerolle F. (2003): Porosity and fluid characterization of granite by capillary wetting using X-ray computed tomography. In: Mees F. *et al*. (eds.): *Applications of X-ray Computed Tomography in the Geosciences.* The Geological Society of London, Special Publication, 215: 95-105.

Hadermann, J., Rösel, F. (1985): *Radionuclide chain transport in inhomogeneous crystalline rocks: limited matrix diffusion and effective surface sorption*. Nagra Technical Report NTB 85-40, Nagra, Wettingen, Switzerland.

Heath, M.J., Montoto, M., Rodríguez-Rey, A., Ruiz de Argandoña, V.G., Menéndez, B. (1992): Rock matrix diffusion as a mechanism of radionuclide retardation: A natural analogue study of El Berrocal granite, Spain. *Radiochimica Acta*, 58/59: 379-384.

Hellmuth, K-H., Siitari-Kauppi, M., Lindberg, A. (1993): Study of porosity and migration pathways in crystalline rock by impregnation with ^{14}C-polymethylmethacrylate. *Journal of Contaminant Hydrology*, 13: 403-418.

Hellmuth, K.H., Klobes, P., Meyer, K., Röhl-Kuhn, B., Siitari-Kauppi, M., Hartikainen, J., Timonen, J. (1995): Matrix retardation studies: size and structure of the accessible pore space in fresh and altered crystalline rock. *Zeitschrift für Geologische Wissenschaften,* 23: 691-706.

Hellmuth, K.-H., Siitari-Kauppi, M., Klobes, P., Meyer, K., Goebbels, J. (1999): Imaging and analyzing rock porosity by autoradiography and Hg-porosimetry/X-ray computertomography-applications. *Physics and Chemistry of the Earth (A)*, 24-7: 569-573.

Ivanovich, M., Longworth, G., Wilkins, M.A., Hasler, S.E. (1987a): *A study of uranium series disequilibrium in core profiles and mineral separated from the samples of Lac du Bonet granite from the URL site, Pinawa, Manitoba, Canada*. Harwell Report, AERE G 4307.

Ivanovich, M., Longworth, G., Wilkins, M.A., Hasler, S.E. (1987b): *A study of uranium series disequilibrium in core profiles and mineral separates from the URL site, Pinawa, Manitoba, Canada*. Harwell Report AERE G 4308.

Ivanovich, M., Harmon, R.S. (1992): Uranium-series Disequilibrium. *Applications to Earth, Marine, and Environmental Sciences*. Oxford Science Publications. 910 p.

Ivanovich, M., Hernández-Benítez, A., Chambers, A.V., Hasler, S.E. (1994): Uranium series study of fracture in materials from El Berrocal site, Spain. *Radiochimica Acta,* 66/67: 485–494.

Johansson, H., Siitari-Kauppi, M., Skalberg, M., Tullborg, E.L. (1998): Diffusion pathways in crystalline rock: examples from Äspö-diorite and fine grained granite. *Journal of Contaminant Hydrology*, 35: 41-53.

Johns, R.A., Steude, J.S., Castanier, L.M., Roberts, P.V. (1993): Non-destructive measurements of fracture aperture in crystalline rock cores using X-ray computed tomography. *Journal of Geophysics Research*, 98-B2: 1889-1900.

Karakan, C.O., Grader, A.S., Halleck, P.M. (2003): Evaluation of local porosity changes in limestone samples under triaxial stress field by using X-ray computed tomography. In: Mees, F., Swennen, R., Van Geet, M., Jacobs, P. (eds.): *Applications of X-ray computed tomography in the geosciences.* Geological Society, London, Special publications 215: 177-189.

Kamineni, D.C., Ticknor, K.V., Vandergraaf, T.T. (1986): Occurrence, composition and radionuclide sorption characteristics of illite from a fractured granite pluton, southeastern Manitoba, Canada. *Clay Minerals*, 21: 909-924.

Kelokaski, M., Siitari-Kauppi, M., Montoto, M., Mateos, F., Martinez-Nistal, A. (2006): fractographic evaluation of low permeable granitic rocks under CLSM and PMMA method from cm to mm scale. *1st. Annual Workshop Proceedings 6TH EC FP - FUNMIG IP*, Saclay (France): p. 6.

Ketcham, R.A., Carlson, W.D. (2001): Acquisition, optimization and interpretation of X-ray computed tomographic imagery: Applications to the geosciences. *Computers and Geosciences*, 27: 381-400.

Kowallis, B., Wang, H.F. (1983): Microcrak study of granitic cores from Illinois Deep Borehole UPH3. *Journal of Geophysial Research*, 88: 7373-7380.

Martínez-Nistal, A. (1993): *Discriminación, cuantificación y cartografiado de componentes petrográficos mediante proceso digital de multi-imágenes microscópicas*. Tesis doctoral. Facultad de Geología. Universidad de Oviedo. (Oviedo, Spain): 235 p.

Mazurek, M. (2000): Geological and hydraulic properties of water-conducting features in crystalline rocks. In: Stober, I. and Bucher, K (eds): *Hydrogeology of crystalline rocks*: 2-3. Kluwer Scientific Publishers, Dordrecht.

Mees, F., Swennen, R., Van Geet, M., Jacobs, P. (2003): Applications of X-ray Computed Tomography in the Geosciences. In: Mees, F., Swennen, R., Van Geet, M., Jacobs, P. (eds): *Applications of X-ray Computed Tomography in the Geosciences*. Geological Society, London, Special Publications, 215, 1-6.

Ménager, M.T., Petit, J.C., Brocander, A. (1982) : The migration of radionuclides in granite: a review based on natural analogues. *Applied Geochemistry. Suppl.* 1: 217-238.

Ménager, M.T., Health, M.J., Barillon, R., Ivanovich, M. (1994): Migration of Uranium from the U-mineralised fractures to the rock matrix in granite: implications for radionuclide transport around a radioactive waste repository. *Proceedings of the International Conference of the Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere, Migration 93*. (Charleston, SC, USA).

Menéndez, B. (1992): *Cuantificación de componentes petrográficos mediante técnicas de microscopía y estereología*. Tesis doctoral. Facultad de Geología. Universidad de Oviedo. (Oviedo, Spain): 130 p.

Miller, W., Alexander, R., Chapman, N., McKinley, I., Smellie, J. (1994): Natural Analogue Studies in the Geological Disposal of Radioactive Wastes. *Studies in Environmental Science*, 57. Elsevier. 395 pp.

Miller, W., Alexander, R., Chapman, N., McKinley, L., Smellie, J. (2000): *Geological disposal of radioactive wastes and natural analogues*. Pergamon. 316 p.

Montoto, M. (2003): *Petrophysics at the rock matrix scale: hydraulic properties and petrographic interpretation*. ENRESA Madrid, Technical Report 03-11: 297 p. ISSN:1134-380X.

Montoto, M.; Montoto, L.; Roshoff, K., Leijon, B. (1980): Microfractographic study of heated and non-heated Stripa granite. *Subsurface Space: Rockstore'80,* 3: 1357-1368. Pergamon Press.

Montoto, M., Heath, M. J., Rodríguez-Rey, A., Ruiz de Argandoña, V. G., Calleja L., Menéndez, B. (1992): Natural analogue and microstructural studies in relation to radionuclide retardation by rock matrix diffusion in granite. *Commission of the European Communities. Nuclear Science and Technology*. EUR 14352 EN: 124 p. ISBN 92-826-4961-X.

Montoto, M., Martínez-Nistal, A., Rodríguez-Rey, A., Fernández-Merayo, N., Soriano, P. (1995): Microfractography of granitic rocks under Confocal Laser Scanning Microscopy. *Journal of Microscopy*, 177(2): 138-149.

Montoto, M. (1996b): Microestructura de rocas y retardo de radionucleidos. *Estratos*, 40: 12-17.

Montoto, M., Ménager, M.T., Rodríguez-Rey, A., Menéndez, B., Martínez-Nistal, A., Fernández-Merayo, N. (1996c): Uranium transfer phenomena in rock matrix: petrophysical and geochemical study of El Berrocal experimental site, Spain. *Journal of Contaminant Hydrology*, 21: 35-46.

Montoto, M. (1999a): Petrophysics: the Petrographic Interpretation of the Physical Properties of the Intact Rock. In: Gottinger Arbeiten zur Geologie und Paläontologie Sb 4. S 210, Special Issue, Textures and Physical Properties of Rocks. ISSN 0534-0403. pp. 132-133.

Montoto, M., Rodríguez-Rey, A., Martínez-Nistal, A., Díez, I. (1999b): *Fractographic characterization by quantitative microscopy of the excavation disturbance caused by boring of the experimental full scale deposition holes in the Research Tunnel at Olkiluoto (Finland)*. Posiva Working Report 99-69 (POSIVA OY Ed., Finland). ISBN 951-652-008-1, ISSN 1239-3096 (EN) 76pp.

Montoto, M., Rodríguez-Rey, A., Martínez-Nistal, A., Díez, I. (1999c): *Fractographic characterization by quantitative microscopy of the excavation disturbance caused by boring of the experimental full scale deposition holes in the Research Tunnel at Olkiluoto (Finland)*. Posiva Working Report 99-69 (POSIVA OY Ed., Finland): p. 76. ISBN 951-652-008-1, ISSN 1239-3096 (EN):

Montoto, M., Rodríguez Rey, A., Martínez Nistal, A., Díez Sarriá, I. (1999d): *Fractographic characterization by quantitative microscopy of the excavation disturbance caused by boring of the experimental full scale deposition holes in the research tunnel at Olkiluoto*. Äspö Hard Rock Laboratory. SKB International Progress Report IPR-01-14. 74p.

Montoto, M. (1999e): *Caracterización petrofísica de las vías de migración de agua en la roca matriz de la arenisca silicificada de Bagombé (Gabón).* Technical Report to ENRESA, 81 p.; Dept. of Geology, Group of Petrophysics, University of Oviedo (Oviedo, Spain).

Montoto, M. (1999f): *Caracterización petrofísica de las vías de migración de agua en la roca matriz del granito pegmatítico de Palmottu (Finlandia).* Technical Report to ENRESA, 87 p.; Dept. of Geology, Group of Petrophysics, University of Oviedo (Oviedo, Spain).

Montoto, M. (2000a): Petrophysical Characterization of the Groundwater Migration Pathways through the Bagombé (Gabon) Rock Matrix Silicified Sandstone. In: Oklo Working Group. Proceedings of the Third and Final Oklo Phase II (ISBN 92-828-9220-4), Michaud, L., von Maravic EUR 19137 EN pp. 273- 282.

Montoto, M. (2000b): Métodos petrofísicos de caracterización de las vías de migración. *IV Jornadas de Investigación y Desarrollo Tecnológico en Gestión de Residuos Radioactivos.* ENRESA (Madrid, España), pp. 40-40.

Neretnieks, I. (1980). Diffusion in the rock matrix: an important factor in radionuclide migration? *Journal of Geophysical Reserch*, 85: 4379-4397.

Pérez del Villar, L., de la Cruz, B., Pardillo, J., Cózar, J.S., Ménager, M.T. (1994): Uranium distribution in relation to the mineralogical and geochemical characteristics of the granite from the El Berrocal uranium mine (Spain). In: *Proceedings of the 4th International Conference of the Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere* (Charleston, SC USA). 363-375.

Pérez del Villar, L., Cózar, J.S., de la Cruz, B., Fernández, A.M. (1995): Influence of the alteration processes on the U-Th-REE minerals in the El Berrocal granite / U-bearing quartz vein system (Spain): an example of natural analogue processes. In: J. Pascua, B. Krobek, K. Zask (eds.): *Minerals Deposits: from their origin to their environmental impacts.* 689-692.

Pérez del Villar, L., Cózar, J.S., Pardillo, J., Pelayo, M., Labajo, M.A. (2000): Lithological and mineralogical analysis of the borehole R-385 (386) core from the Palmottu site (Finland). In: *CIEMAT Contribution to "Transport of radionuclides in a natural system at Palmottu (Finland). Phases I and II*. Compiled by L. Pérez del Villar. CIEMAT/DIAE/54321/5/00.

Perret, J., Prasher, S.O., Kantzas, A., Langford, C. (1999): Three-dimensional quantification of macropore networks in undisturbed soil cores. *Soil Science Society of American Journal*, 63: 1530-1543.

Petford, N., Miller, J.A. (1990): SLM confocal microscopy: an improved way of viewing fission tracks. *Journal of the Geological Society*, 147: 217-218.

Petford, N., Miller, J.A. (1992): Three dimensional imaging of fission tracks using confocal scanning laser microscopy. *American Mineralogist*, 77: 529-533.

Petford, N., Miller, J.A. (1993): The study of fission tracks and other crystalline defects using confocal scanning laser microscopy. *Journal of Microscopy*, 170: 201-212.

Pirhonen, V. (1990): *Porosity and focussed dissolution of granitic rocks in two study areas in southern Finland. Aspects of methodology*. Tech. Res. Centre of Finland (VTT). Publ. 69. Espoo, Finland.

Pyralk-Nolte, L.J., Montamagno, C.D., Nolte, D. (1997): Volumetric imaging of aperture distributions in connected fracture networks. *Geophysical Research Letters*, 24: 2343-2346.

Rasilainen, K., Hellmuth, K.H., Kivekäs, L., Melamed, A., Ruskeeniemi, T., Siitari-Kauppi, M., Timonen, J., Valkiainen, M. (1996): *An interlaboratory comparison of methods for measuring rock matrix porosity.* VTT Research Notes 1776, Technical Research Centre of Finland, Espoo, Finland.

Ruiz de Argandoña, V.G., Rodríguez-Rey, A., Celorio, C., Calleja, L., Suárez del Río, L.M. (2003): Characterization by X-ray computed tomography of water absorption in a limestone used as a building stone in the Oviedo Catedral (Spain). In: Mees, F., Swennen, R., Van Geet, M., Jacobs, P. (eds): *Applications of X-ray Computed Tomography in the Geosciences*. Geological Society, London, Special Publications, 215: 127-134.

Schild, M. (1999): *Verbundene Mikroporositäten in Kristallingesteinen-Fallstudie Felslabor Grimsel*. PhD Thesis, University of Göttingen, Göttingen.

Schild, M., Siegesmund, S., Vollbrecht, A., Mazurek, M. (2001): Characterization of granite matrix porosity and pore-space geometry by in situ and laboratory methods. *Geophysical Journal International,* 146: 111-125.

Siitari-Kauppi, M. (1995): *Investigation of Porosity and Microfracturing in a Disturbed Zone with the ^{14}CPMMA Method Based on Samples from Full-Scale Experimental Deposition Holes of the TVO Research Tunnel, Nuclear Waste Commission of Finnish Power Companies*. Report YJT-95-13.

Siitari-Kauppi, M., Flitsiyan, E.S., Klobes, P., Meyer, K., Hellmuth, K.H. (1998a): Progress in physical rock matrix characterization: Structure of the pore space. Materials Research Society. Symposium Proceedings v 506. *Scientific Basis for Nuclear Waste Management*. Warrendale, PA, USA. p 671-678.

Siitari-Kauppi, M., Flitsiyan, E.S., Klobes, P., Meyer, K., Hellmuth, K.-H. (1998b): Progress in physical rock matrix characterization: structure of the pore space. In: I.G. McKinley, C. McCombie (edits.): *Scientific Basis for Nuclear Waste Management XXI*. Mat. Res. Soc. Symp. Proc. 506.

Simmons, D., Richter, D. (1976): Microcraks in Rocks. In: Strens, R.G.J. (Ed.). *The Physics and Chemistry of Minerals and Rocks*. John Wiley & Sons Ltd.: 105-137.

Sugawara K., Fukahori D., Iwatani T., Kubota S. (2004): Analisis of water flow in rock by a Tracer-aided X-ray CT. In: Otani, J., Obara, Y. (ed.): *X-ray CT for Geomaterials. Soils, Concrete, Rocks*. Balkema Publ.: 335-344.

Ticknor, K.V., Vandergraaf, T.T., Kamineni, D.C. (1986a): *Radionuclide sorption on mineral and rock thin sections. Part II: Sorption on granitic rock*. Geochemistry and Applied Chemistry Branch, Whiteshell Nuclear Research Establishment, Atomic Energy of Canada Ltd. TR-385. Pinawa, Manioba (Canada), R0E 1L0. June1986.

Ticknor, K.V., Vandergraaf, T.T. (1986b): A method for producing autoradiographs of rough surfaces. *Nuclear and Chemical Waste Management*, 6: 233-239.

Vandergraaf, T.T., Abry, D.R.M., Davis, C.E. (1982): The use of autoradiography in determining the distribution of radionuclides sorbed on thin sections of plutonic rocks from the Canadian shield. *Chemical Geology*, 36: 139-154.

Van Geet, M., Swennen, R., David, P. (2001): Quantitative coal characterisation by means of microfocus X-ray computer tomography, colour image analysis and back scatter scanning microscopy. *International Journal of Coal Geology*, 46: 11-25.

Van Geet, M., Lagrou, D., Swennen, R. (2003): Porosity measurements of sedimentary rocks by means of microfocus X-ray computed tomograpy (μCT). In: Mees, F., Swennen, R., Van Geet, M., Jacobs, P. (eds): *Applications of X-ray Computed Tomography in the Geosciences*. Geological Society, London, Special Publications, 215: 51-60.

Vervoort A., Van Geet M., Sellers E. (2004): Recent advances of X-ray CT and its applications for rock material. In: Otani, J., Obara, Y. (ed.): *X-ray CT for Geomaterials. Soils, Concrete, Rocks.* Balkema Publ.: 79-91.

Yoshida, H., Kodama, K., Ota, K. (1993): Role of microscopic flow-paths on nuclide migration in sedimentary rocks: a case study from the Tono uranium deposit, central Japan. *Proceedings of the 4th International Conference of the Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere, Migration 93.*

Yoshida, H., Kodoma, K., Ota, K. (1994): Role of microscopic flow-paths on nuclide migration in sedimentary rocks. A case study from the Tono uranium deposit.Central Japan. *Radiochimica Acta,* 66/67: 505–511.