Äspö Hard Rock Laboratory

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SKB

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This report concerns a study which was conducted for SKB. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

USE OF THE ^{14}C-PMMA AND HE-GAS METHODS TO CHARACTERISE EXCAVATION DISTURBANCE IN CRYSTALLINE ROCK

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ABSTRACT

Characterisation of the excavation disturbance caused by boring of experimental full-scale deposition holes in the Research Tunnel at Olkiluoto was carried out successfully by using two novel methods; the ^{14}C-PMMA and He-gas methods, which were modified and applied for the first time in this type of study. The experience obtained implies that the techniques are feasible and can be used to study similar types of rock excavation disturbance such as that caused by boring with mini discs, a technique which will be used in the underground Hard Rock Laboratory at Äspö during late 1998 and early 1999. Both of the measurement methods have been in continuous use and the work has included development of both the measuring and interpretation techniques. Use of the ^{14}C-PMMA method is suggested for studies of rock structure and the spatial distribution of porosity. The ^{14}C-PMMA method also provides quantitative information about nanometre-range porosity which is beyond the scope of most standard methods of microscopic investigation. The use of He-gas methods are proposed for determining the diffusion coefficient, permeability and complementary porosity of rock samples taken from the disturbed zone.

SAMMANFATTNING

Karakterisering av den borrstörda zonen i de simulerade, fullstora deponeringshålen i forskningstunneln i Olkiluoto har genomförts med lyckat resultat genom användning av två nya metoder: ^{14}C-PMMA- och He-gas metoderna, vilka har applicerats för första gången i denna typ av studie. Den erfarenhet som erhållits indikerar att teknikerna är möjliga att använda och att de kan utnyttjas för liknande studier av den störda zonens egenskaper till exempel vid borrning med minidiskar, som skall tillämpas under jord i Äspölaboratoriet under 1998/1999. Båda mätmetoderna har använts vid upprepade tillfällen och arbetet har inkluderat utveckling av både mätteknik och utvärderingssätt. ^{14}C-PMMA-metoden föreslås användas till studier av bergets struktur och rumsliga fördelning av porositet. ^{14}C-PMMA–metoden lämnar även kvalitativ information om porositet i nanometerskala, vilket är en större upplösning än de flesta mikroskopiska standardmetoder. He-gasmetoden föreslås användas för bestämning av diffusionskoefficient, permeabilitet och porositet hos bergprover tagna från den störda zonen.

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1 INTRODUCTION

Characterisation of the excavation disturbance caused by boring of the experimental full scale deposition holes in the Research Tunnel at Olkiluoto was carried out successfully in 1995-1997 (Autio & Kirkkomäki 1996, Autio 1996). Studies of disturbance were carried out in the laboratory by examining 98 mm diameter core samples representing different boring parameters taken from different locations in the full-scale holes.

The properties of both disturbed and undisturbed rock were investigated. Novel ^{14}C-PMMA and He-gas methods were modified and applied for the first time in this type of study. The methods were found to be useful and to provide new information about the properties of the disturbed zone.

The ^{14}C-PMMA method was found to be a practical and efficient tool for studying rock structure and the spatial distribution of porosity. The method effectively fills the gap between macroscopic and microscopic investigation methods. In addition, it provides quantitative information about nanometre-range porosity which is beyond the scope of most standard methods of microscopic investigation.

The He-gas method proved useful when determining the diffusion coefficient and permeability of large rock samples. Permeability can be measured by using relatively low pressure gradients and low flow velocities. The diameter of the samples (98 mm) was approximately one hundred times larger than the rock grain size and included a large number of structural features which could have hampered measurements and caused significant variation in the results obtained if a smaller sample size had been employed. There is no other experimental method currently available which could be used to determine the properties of this size of samples in the same time period.

The experience gained in characterisation studies of the excavation disturbance caused by the boring of experimental full-scale holes in the Research Tunnel at Olkiluoto imply that the measurement techniques are practical and feasible. They are therefore considered suitable for use when studying a similar type of excavation disturbance such as that caused by boring using mini discs, a process which is planned to be used in the underground hard rock laboratory at Äspö during the later half of 1998 and early 1999.

As a proposal for how the methods should be used at Äspö, an updated description of the techniques, based on experience which has not yet been fully reported, is presented in this report.

The principles behind both methods and their relevant applications have been well documented previously and the reader is advised to look for complementary information on details from the following sources. The principles of the ^{14}C-PMMA method are presented in (Hellmuth et al. 1993, Hellmuth et al. 1994) and its recent application in characterisation of the disturbed zone have been presented in (Siitari-Kauppi 1995, Autio 1996, Autio & Siitari-Kauppi 1997, Autio et al. 1998, Siitari-Kauppi et al. 1995, Siitari-Kauppi & Autio 1997a and Siitari-Kauppi & Autio 1997b).

The principles of the He-gas method have been presented in (Hartikainen et al. 1994, Hartikainen et al. 1996); and its recent applications in characterisation of the disturbed zone have been presented in (Hartikainen et al. 1995, Autio 1996, Autio et al. 1998).

The coring of the 98 mm diameter samples required for laboratory sample preparation, mapping, notation and subsequent documentation can be carried out in several ways. The techniques employed in the Research Tunnel at Olkiluoto have been presented in (Autio et al. 1995, Autio et al. 1996). A sample diameter between 90 and 100 mm is considered appropriate, since it offers good coverage of the exposed rock surface and is still relatively easy to handle during sample preparation.

2 ^{14}C-PMMA METHOD

2.1   INTRODUCTION

The ^{14}C-PMMA method makes it possible to study the spatial distribution of the pore space in rock and the heterogeneity of rock matrices at the submicrometre to centimetre scale. Subsequent autoradiography and digital image analysis make it possible to analyse features limited in size by the range of ^{14}C beta radiation.

The ^{14}C-PMMA method involves impregnation of centimetre-scale rock cores with ^{14}C-labelled methylmethacrylate (^{14}C-MMA) in a vacuum, irradiation polymerisation, sample partitioning, autoradiography, optical densitometry and porosity calculation routines (see Figure 2-1) using digital image processing techniques. The technique of impregnation with the labelled low-molecular-weight and low-viscosity monomer ^{14}C-MMA, which wets silicate surfaces well and can be fixed by irradiation polymerisation, provides information about the accessible pore space in crystalline rock that cannot be obtained by using other methods.

Total porosity is calculated by employing 2D autoradiographs of sawn rock surfaces. The geometry of porous regions is then visualised. The preconditions for applying this method are: (i) a known local bulk rock density; (ii) the presence of only two phases - mineral and PMMA; and (iii) the homogeneous distribution of pores and minerals below the limit of the lateral resolution of autoradiography. Fissures and cracks with apertures larger than 20 μm are usually excluded from the quantitative measurements.

In addition to the ^{14}C-PMMA method complementary microscopy, scanning electron microscopy (SEM) and Back Scattered Electron (BSE) images are also used to make qualitative investigations of the pore apertures and minerals in porous regions in greater detail.
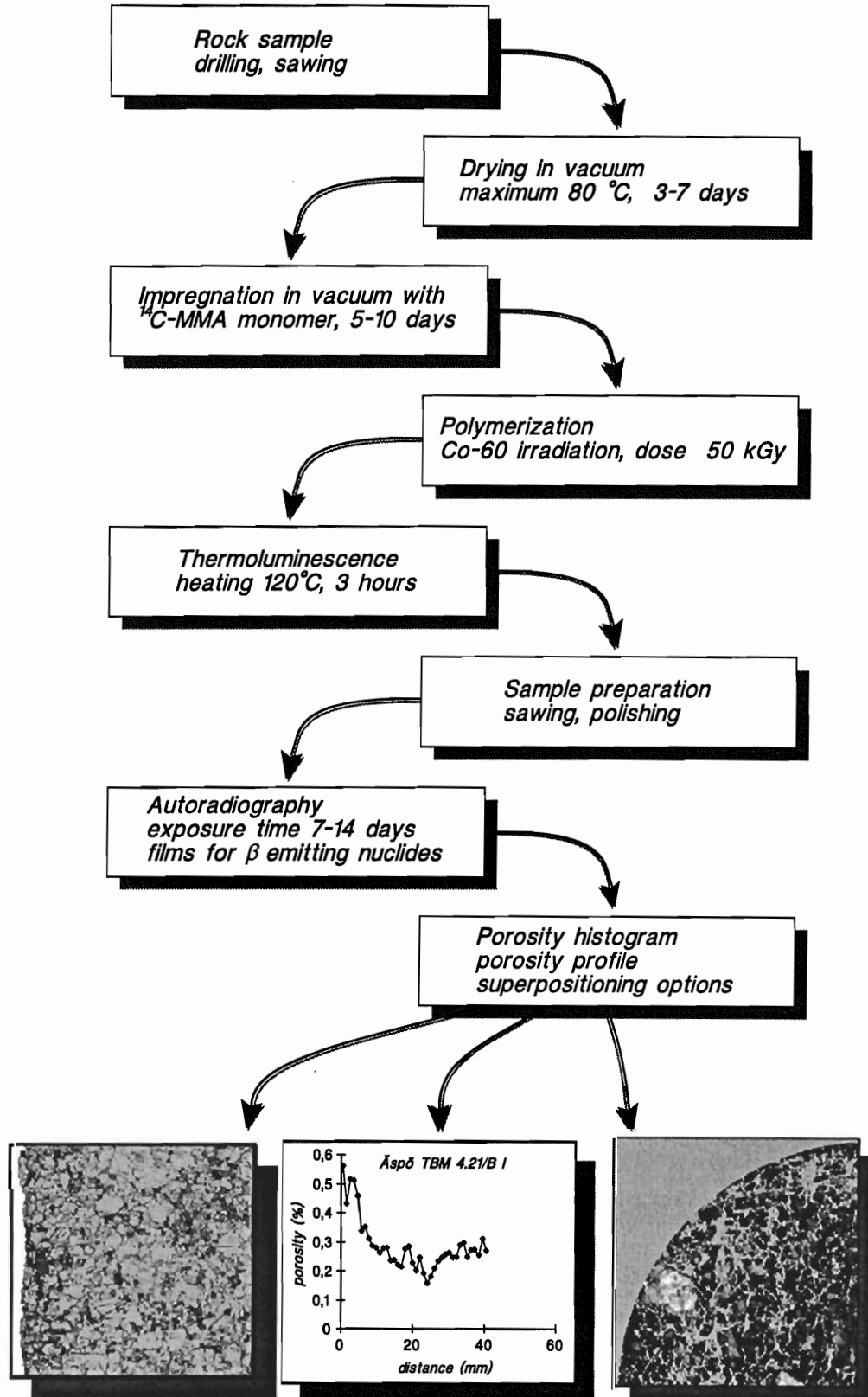
Figure 2-1. *The ^{14}C-PMMA method.*

2.2   SAMPLE PREPARATION

The size of the rock samples fixes the size of the vacuum impregnation chamber. The diameter of the cylindrical vacuum chamber should be as close as possible to the diameter of the rock sample so that the gap between the chamber wall and the rock sample is as small as possible. Vacuum chambers made for 98 and 95 mm diameter samples already exist and therefore use of this sample diameter is suggested. If other diameters are employed, it is wise to select them according to the diameter of the tubing from which the vacuum chambers will be made. The length of samples should be large enough to ensure that a part of the sample represents undisturbed rock. According to the results of investigations on samples from the TBM-tunnel at Äspö and from the full-scale deposition holes in the Research Tunnel, a sample length of about 40 mm has been found to be appropriate.

Each rock sample is partitioned in different steps during the course of the investigation. A schematic presentation of the original position of the core sample in the full-scale deposition hole and the sawing procedures used when partitioning the sample is shown in Figure 2-2.

Sample partitioning is carried out in two phases. The dashed line in Figure 2-2 divides the procedure into partition before impregnation (above the dashed line) and after impregnation (below the dashed line).

Before impregnation a 40 mm thick section of the original core sample is sawed to be impregnated. Partition following impregnation is carried out for autoradiography, scanning electron microscopy (SEM) and petrologic studies of thin sections by using polarisation microscopy. The shaded surfaces in Figure 2-2 are exposed to autoradiographic film, several autoradiographs being taken. Partitioning after sample impregnation is flexible and can be carried out in steps based on the results obtained.

The studied rock surfaces used to produce autoradiographs are finished after sawing by grinding and polishing on a grinding plate using a fine grinding powder to produce artefact-free specimens. Suitable grain numbers for the grinding powder are 180, 220, 400 and 600 in steps. The objective of the grinding is to improve the resolution of the autoradiographs and also to ensure that the rock surfaces are free of artefacts resulting from possible contamination by the impregnant during the sawing process.
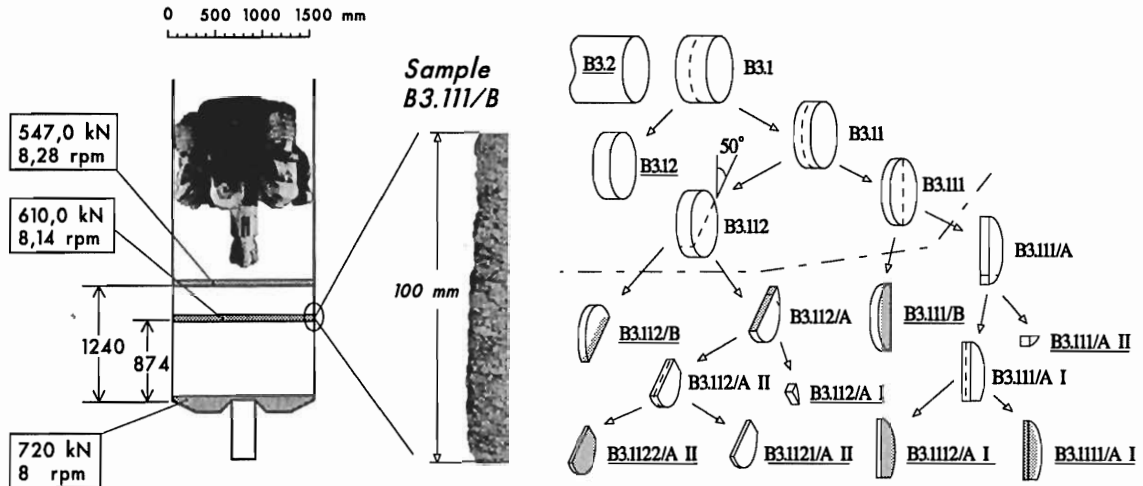
Figure 2-2. Partitioning of a 98 mm diameter rock core sample. Surfaces exposed during the 14C-PMMA investigation are shaded.

2.3 DRYING AND IMPREGNATION

Rock samples must be dried before impregnation to remove any water which could hamper the penetration of the impregnant into the pore spaces. It is recommended that 8 to 14 days drying under vacuum at a maximum temperature of 80°C is employed, these conditions having been assessed as not causing any significant deformation of microfractures or pore space in the rock. After drying, samples should be allowed to cool slowly to a room temperature of about 18°C under vacuum conditions.

For ^{14}C-MMA impregnation, the tracer is placed in a 50 ml reservoir and transferred under vacuum to the impregnation chamber. Slow transfer of the monomer ensures degassing of the monomer and infiltration of vapour only. Impregnation time ranges from seven to 22 days. The same impregnation times should be used for all samples of the same rock type. After impregnation, the samples are irradiated with gamma rays from a Co-60 source in order to polymerise the monomer in the rock matrix. The required dose is about 50 kGy (5 Mrad). Samples are irradiated in polyethylene vials under a ^{14}C-MMA/water emulsion to prevent diffusion.

Methylmethacrylate (MMA) is a monomer with very low viscosity, 0.00584 P (20°C) (Daniels & Alberty 1967), while the viscosity of water is 0.00895 P (25°C). Because its contact angle with silicate surfaces is low, the impregnation of bulk rock specimens by MMA is rapid and related to existing pore apertures. The MMA molecule is small (mol.weight 100.1). It has non-electrolytic properties and only low polarity, the polarity of the ester being considerably lower than that of water, and its behaviour in the rock

matrix is that of a non-sorbing tracer. The low β energy of the ^{14}C isotope, a maximum of 150 keV, is convenient for autoradiographic measurements.

The monomer used is ^{14}C-labelled MMA with a specific activity of 2-5 mCi/g and a total activity of 50 mCi. Its radiochemical purity is >95%. In this application the dilution of the tracer usually ranges between 1 850 000 Bq/ml (50 μCi/ml) and 925 000 Bq/ml (25 μCi/ml). The tracer activity used is determined after every impregnation by using liquid scintillation counting (Rackbeta 280). The calibration sources are prepared by diluting the ^{14}C-labelled MMA with inactive MMA. The resulting activities typically range from 462 Bq/ml (12.5 nCi/ml) to 185 000 Bq/ml (5 μCi/ml).

2.4 AUTORADIOGRAPHY

After impregnation and irradiation polymerisation, autoradiography of the samples is performed. Examples of autoradiographs of rock sections are shown in Figure 2-3 and 2-4.

Irradiation of rocks with Co-60 causes strong thermoluminescence of K-feldspar and other major rock-forming minerals, which would result in exposure of autoradiographic film. To avoid this, the thermoluminescence is released by heating the irradiated samples to 120°C for 3 hours before they are sawn.

After cooling, the samples are sawn as shown in Figure 2-2. The sawn rock surfaces are exposed after grinding on Kodak BioMax MS autoradiographic film which is a high-performance autoradiographic film for ^{14}C and other low energy β-emitting nuclides. The resolution of β film is a few μm. Mylar foil with an aluminium coating was placed on top of the film to shield it from other emissions caused by luminescence.

The final resolution of the autoradiograph depends on the roughness of the sawn surface and the range of the 150 keV beta particles in the rock matrix. As to the penetration range of beta radiation, the rock samples used are considered to be infinite in thickness. The beta absorption correction is obtained from the ratio of the densities of rock and the MMA.

With the level of tracer activity and the autoradiographic films employed, exposure times for samples typically lie between 9 and 24 days. The same exposure times should be used for samples of the same rock type to obtain results which are as technically comparable as possible. Final exposure times should be decided after a first test on the basis of features of interest. If there is a great difference in the porosity of the disturbed and undisturbed rock it is suggested that two different exposure times are used.
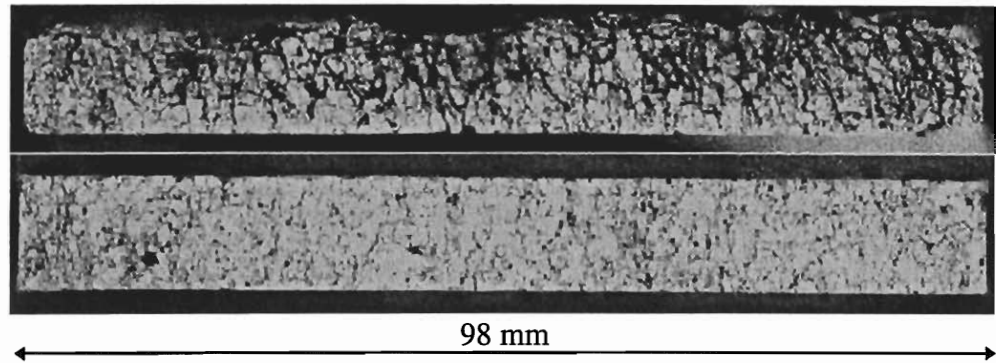
Figure 2-3. *Autoradiographs of disturbed rock, sample B3.111/B (top) and undisturbed rock, sample B3.112/B (bottom). The section shown is perpendicular to the disturbed surface. The gap between the sections, which are on the same plane, is 1.5 mm.*

Figure 2-4. *Examples of autoradiographs of sections from the surface of the full scale deposition holes produced with the novel ^{14}C-PMMA method. Different levels of grey shading represent different porosity values.*

Accurate determination of the profile of the disturbed surface has been hampered by the β-radiation emissions from the free sample surface which results in an inaccuracy of about 0.5 mm in the depth of the porosity profiles. It is suggested that radiation shielding be used in future measurements to prevent these emissions. Suc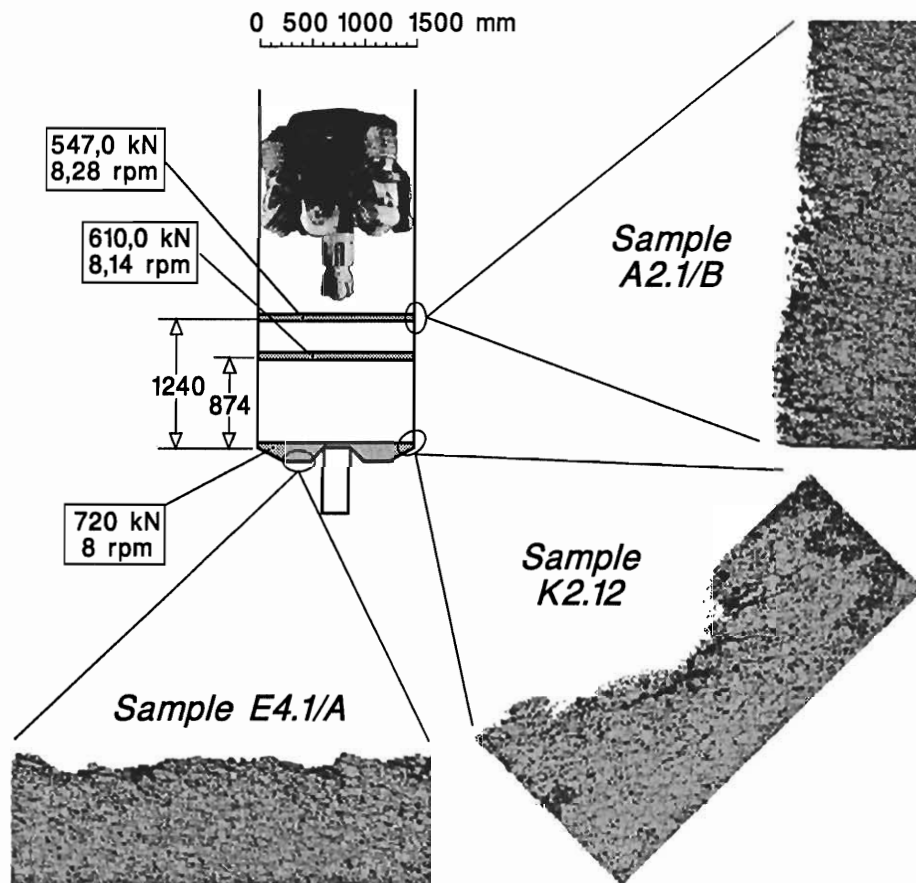h shielding can be made by sealing the free sample surface with a material which absorbs β-radiation before the autoradiography is performed. Plenty of suitable materials are available such as a thin layer of epoxy or some soft plastic material such as a wax.

2.5   DIGITAL IMAGE PROCESSING

2.5.1   General

Once obtained, the autoradiographs are scanned to obtain a digital image by using a high-resolution table scanner in transparent light mode. The scanner must be calibrated to ensure that the greyscale of the images is not altered. Before scanning, all the scanner's image enhancement features must be disabled to normalise the image recording process.

After the digital image has been acquired, it is transformed from an intensity image to an optical density image and later into a porosity image. It is this porosity image which is then further analysed to study the spatial distribution of porosity in selected areas of interest.

Digital image analysis divides the autoradiograph into rectangular units of area called pixels. 300 dpi scanning resolution is used in the analysis which means a pixel size of about 85x85 µm. The intensities found in each subdomain are converted into corresponding optical densities, and these in turn are converted into activities with the help of the calibration curves measured for each exposure. Finally, the activities found are converted into the respective porosities. In principle, image interpretation is based on studying the abundance of tracer in each subdomain.

2.5.2   Intensity and optical density

Since the response of the image source (video camera or table scanner) and the amplifier of the digital image analyser are linear, the digitised grey levels of the film can be treated as intensities. Here, intensity means the quantity of light coming through the autoradiographic film.

Optical densities, which according to Lambert & Beer's law are proportional to concentration, are derived from the intensities as follows:

$$D = \log\left(\frac{I_0}{I}\right)$$ (2.5-1)

where D is the optical density, I_0 is the intensity of the background and I is the intensity of the sample. It can be seen that as the intensity decreases (i.e. the film gets darker), the optical density increases.

2.5.3 Activity and optical density

A conversion function is needed to relate the measured optical densities (grey levels) to corresponding activities. ^{14}C-PMMA standards (tracer diluted with inactive MMA) having specific activities between 462 and 185 000 Bq/ml are used to establish the calibration function. The following calibration curve applies:

$$D = D_{max}\left(1 - e^{-kA}\right)$$ (2.5-2)

where D_{max} is the maximum optical density, k is a fitting parameter, and A is the specific activity. Solving for A from Eq. 2.5-2 gives:

$$A = -k^{-1}\ln(1 - D / D_{max})$$ (2.5-3)

2.5.4 Porosity

The local porosity ε of the sample was simply obtained from the abundance of the tracer (i.e. assuming constant tracer concentration in the PMMA, the higher the abundance of the tracer, the higher the local porosity):

$$\varepsilon = \beta(A / A_0)$$ (2.5-4)

where A_0 is the specific activity of the tracer used to impregnate the rock matrix, and β is the β-absorption correction factor. The absorption of β radiation in a substance depends in an approximately linear fashion on the density of the substance. Factor β can therefore be approximated as follows:

$$\beta = \rho_s / \rho_0$$ (2.5-5)

where ρ_s is the density of the sample and ρ_0 is the density of pure PMMA (1.18 g/cm^3). In this interpretation the sample is assumed to consist of rock

material and pores (which contain PMMA), and therefore ρ_s can be expressed as:

$$\rho_s = \varepsilon\rho_0 + (1-\varepsilon)\rho_r \tag{2.5-6}$$

where ρ_r is the density of the mineral grains. In practice, the average density of the rock sample is used in bulk measurements. Using Eqs. 2.5-5 and 2.5-6 in Eq. 2.5-4, the relationship between porosity and activity can be solved for:

$$\varepsilon = \frac{\dfrac{\rho_r}{\rho_0}\dfrac{A}{A_0}}{1+\left(\dfrac{\rho_r}{\rho_0}-1\right)\dfrac{A}{A_0}} \tag{2.5-7}$$

where A is the specific activity of an individual pixel and A_0 is the specific activity of the tracer.

The porosity of each individual pixel n from the autoradiogram is calculated according to Equations 2.5-3 and 2.5-7. The porosity histogram gives the relative frequency of regions of each individual porosity. The total porosity is obtained from the porosity distribution by taking a weighted average:

$$\varepsilon_{tot} = \frac{\sum\limits_n Area_n \varepsilon_n}{\sum\limits_n Area_n} \tag{2.5-8}$$

where $Area_n$ is the area of pixel n, and ε_n is the local porosity of pixel n.

The amount of tracer in the sample, and the volumetric porosity, can thus be derived from the blackening of the film caused by radiation emitted from the plane surface of the rock section. If the pore sizes are well below the resolution of the autoradiograph, the major fraction of the beta radiation emitted is attenuated by silicate. The tracer can thus be considered to be diluted by the silicate. For the ^{14}C-PMMA method to be used, the bulk density must be known; there must be only two phases (mineral and PMMA), and both pores and minerals must be homogeneously distributed below the lateral resolution limit of the autoradiography. Fissures or cracks with apertures of 20 μm or more are not comparable with the calibration sources.

The sample porosity profiles shown in Figure 2-5 are measured from autoradiographs taken from the surfaces of sawn rock samples. Each profile typically consists of 7 to 15 measurements in which the thickness of the

digitally-scanned sector is between 7 and 14 mm. The total porosity profile of each sample is the arithmetic or weighted average of the measured sectors.
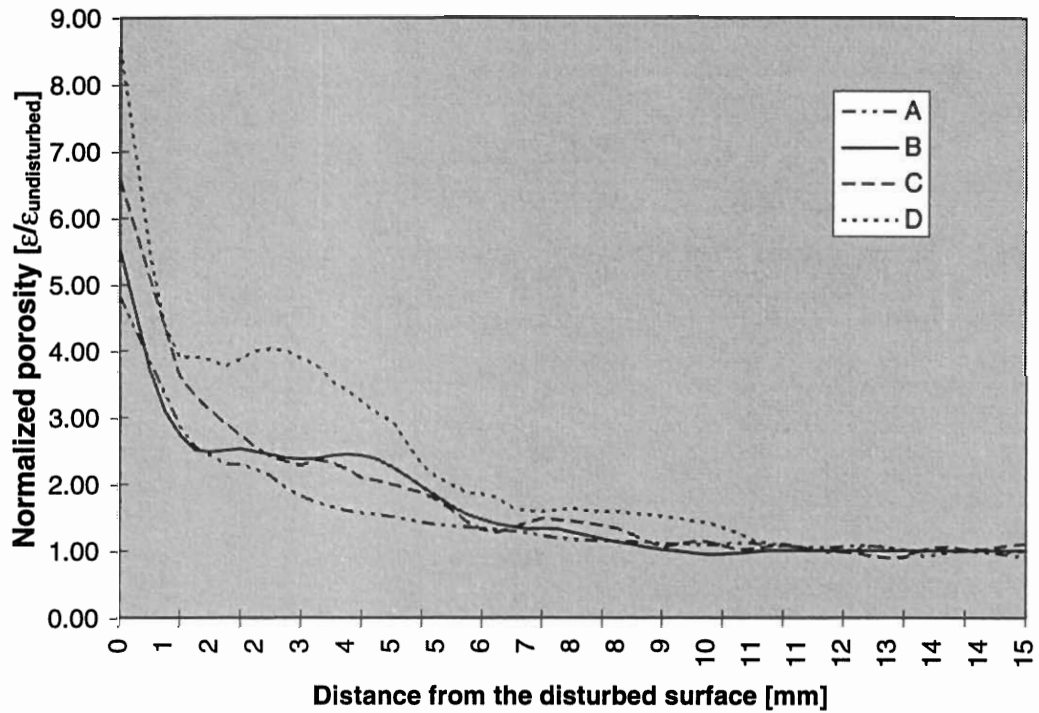

Figure 2-5. Normalised porosity profiles of samples A, B, C and D perpendicular to the wall of the full-scale holes. The average value of the porosity of undisturbed rock is 0.14 vol.%.

3 HE-GAS METHOD

3.1 INTRODUCTION

The He-gas method is used to establish the disturbance of rock in terms of effective diffusion coefficient and permeability. The method is based either on diffusion, see Figure 3-1, or the flow of helium through a rock sample saturated with nitrogen gas. In the case of permeability measurements, flow through the sample is caused by a pressure gradient as shown in Figure 3-2.
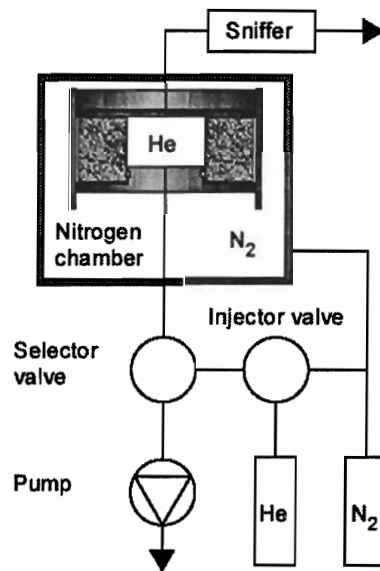
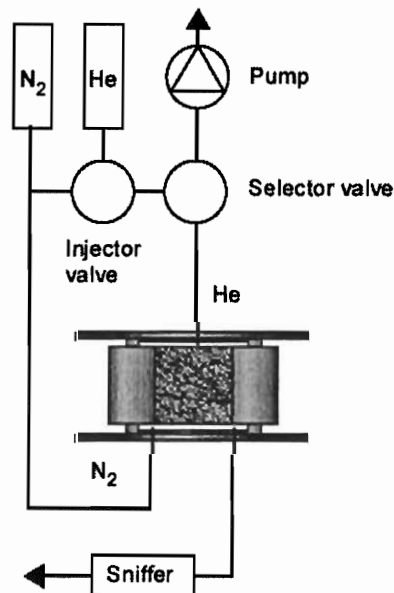*Figure 3-1. Experimental set up for through-diffusion measurements.*

Figure 3-2. Experimental set up for permeability measurements.

The main steps in the procedure are:

1)   Preliminary sample partitioning
2)   Drying of samples
3)   Sealing of the disturbed surface using epoxy
4)   Final sample preparation
5)   Drying
6)   Determination of porosity
7)   Measurement of diffusion coefficient
8)   Measurement of permeability
9)   Partitioning of samples into undisturbed and disturbed sections
10)   Repetition of steps 5-9.

3.2   SAMPLE PREPARATION

Two types of sample geometry are used in the He-gas measurements. Disc samples with a central hole (i.e. rings) are used to measure the average permeability and the diffusion coefficient, see Figure 3-3. The other type of sample is a cube, and these are used to measure properties in two different perpendicular directions. Measurements on the cubes are technically more difficult and experience has shown that more repetition of measurements is required. If the rock material under consideration is homogeneous and not oriented, it is proposed that only disc samples (rings) are used.
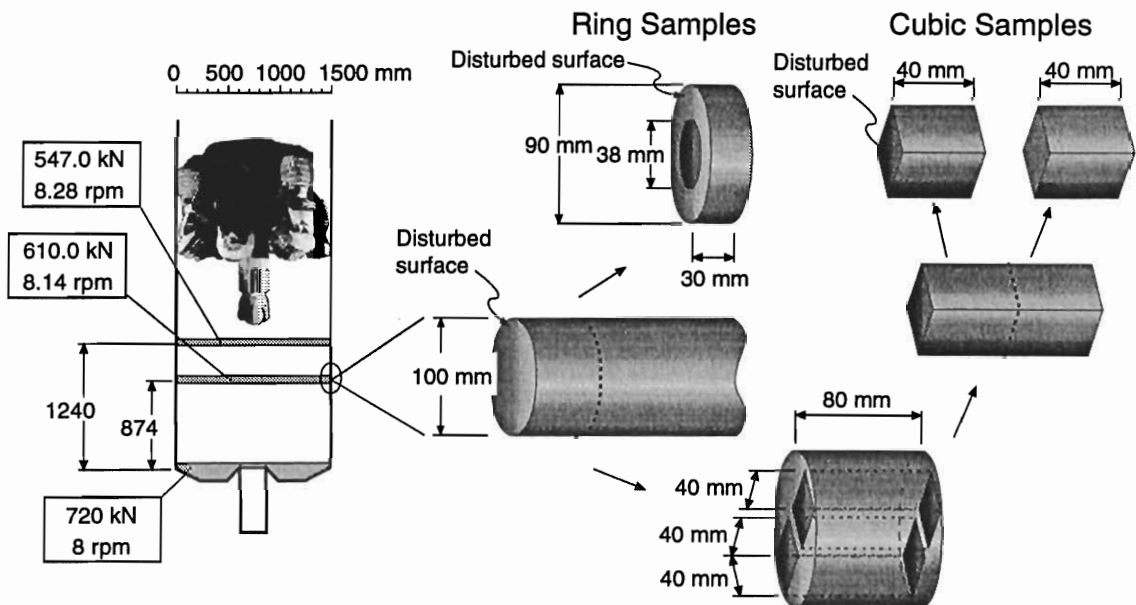

Figure 3-3. The sampling and preparation of ring and cubic samples.

The disturbed surfaces are first sealed by using epoxy before the samples are placed in the measuring chambers and sealed in place using an elastic-plastic sealing material with the brand name Terostaat made by Würth Gmbh.

In both the ring samples and the cubic samples the disturbed surface is sealed with Caldofix fluorescent epoxy before measurements are carried out. The idea behind using the epoxy seal is that sealant will enter the open, large-aperture connected fractures which are close to the disturbed surface and thus prevent rapid flow along these fractures. A similar effect might be achieved in the long term by using compacted bentonite as a sealing material under swelling pressure in saturated conditions. The depth of penetration of bentonite into fractures has been studied experimentally (Pusch 1977 and Pusch 1983), but the penetration of bentonite into disturbed surfaces such as those in the full-scale holes in the Research Tunnel at Olkiluoto has not been established, and therefore the actual similarity of epoxy and bentonite sealings cannot be assessed.

3.2.1   Ring samples

The shape and dimensions of the ring samples from the Research Tunnel at Olkiluoto which were measured by the through-diffusion method are shown in Figure 3-4. The upper side of the sample is the disturbed zone sealed by fluorescent epoxy.

The idea behind using this measurement geometry is to obtain a result which is averaged over all possible lateral directions, and thus independent of any possible orientation effects caused by the structure of the sample.
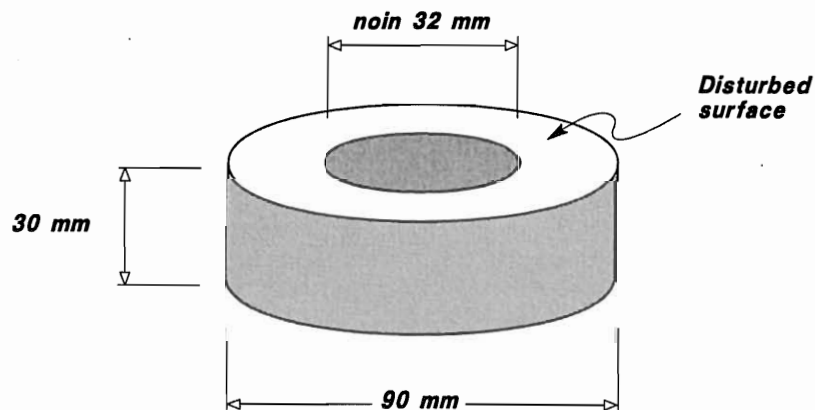
Figure 3-4. Ring sample.

3.2.2 Cubic samples

Cubic samples are sawn from cylindrical core samples as shown in Figure 3-5. After measuring porosity by using the helium pycnometer, samples are cut into two parts as shown in Figure 3-6 so that one part includes the disturbed zone (Samples A) and the other part is intact rock (Samples B).
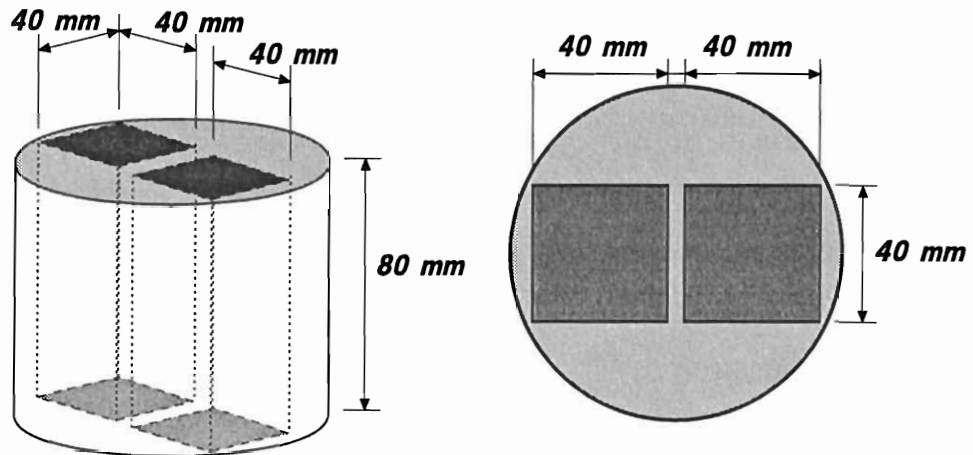

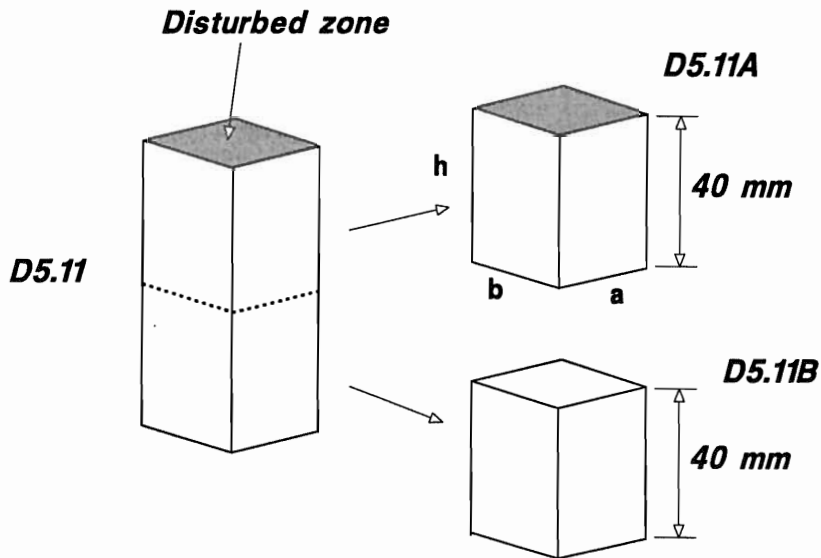*Figure 3-5. Sawing of cylindrical samples to obtain cubic samples.*

Figure 3-6. Further cutting of the cubic samples.

3.2.3   Sealing of the disturbed surface using epoxy

The material used for sealing the disturbed surface was a fast-curing epoxy resin, brand name Caldofix HQ by Struers A/S, which comprises two fluid components. Caldofix has to be heated to at least 70°C to cure and it is characterised by minimal shrinkage. The resin/hardener mixing ratio is 20/6. The pot life of the mixed resin at 20 °C is 4 hours. The curing time at 70°C is 90 minutes. The gelation time when the material starts to polymerise is approximately 50 minutes. Fluorescent dye, brand name EpoDye by Struers, is mixed into the epoxy.

The resin and hardener are mixed at room temperature. Before carrying out the actual sealing of samples, the polymerisation time (assumed here to be 50 minutes) of the specific batch is checked. The sealing procedure after drying of samples is as follows:

1) The sample is heated to 70°C.

2) The dye (1.2 g) is mixed with the hardener (69.5 g).

3) The two components of the epoxy (233 g resin and 69.5 g hardener) are mixed together.

4) The sample and the epoxy are kept in an oven at 70°C for 46 minutes.

5) When 46 minutes have elapsed from the time when the mixing of the epoxy was started, the polymerising mixture is poured onto the disturbed surface of the rock sample so that the layer of epoxy is less than 1 mm thick.

6) The coated sample is kept in the oven at 70°C until the resin is completely cured.

The procedure described here was used for the rock samples taken from the Research Tunnel at Olkiluoto. However, at the time when this report was written, the exact penetration depth of the epoxy sealant into specific rock samples had not been verified. It will be investigated using fluorescent microscopy in a quantitative manner before the end of 1998. If required, the penetration depth of the epoxy can be adjusted by lengthening the settling time or by using different type of polymer.

3.3 THROUGH DIFFUSION MEASUREMENTS

3.3.1 Measuring techniques

In the through-diffusion measurements the injection chamber is first flushed with nitrogen and then evacuated by using a vacuum pump. This is continued until the He-concentration is stabilized at a very low level. After evacuation the pressure of the injection chamber will be less than 0.08 bar.

During the evacuation, helium flows through the injection loop. After the evacuation, loop containing helium is connected to the injection chamber by using a 6-way valve and an injector valve. At the same time the other end of the loop is connected to nitrogen flow that goes through a pressure equalizer. Because of this, only the helium in the loop (5 ml) will be injected. Helium is sucked from the loop to the injection chamber by the vacuum. After connecting the loop to the injection chamber, the system is let to stabilize for two minutes. In that time all helium is transferred to the injection chamber, and the valve is turned off. Helium now diffuses through the sample to the sniffer chamber, which is continuously flushed with nitrogen. The helium concentration of this nitrogen is then measured with a Leybold helium-leak tester. During the through-diffusion, the injection chamber is sealed. The whole through-diffusion measurement takes about 1200 minutes. The purity of the helium used is 99.995% and that of the nitrogen used is 99.9%. It is important in the through-diffusion method that there is no pressure difference between the injection and sniffer chambers, and helium migrates through the sample only by diffusion.

3.3.1.1 Ring samples

In the case of ring samples the helium pulse is injected into the hole at the centre of the sample. The outer cylindrical surface of the sample is then flushed with nitrogen to remove and catch helium molecules that have diffused through the rock and the helium concentration of the nitrogen is then measured. The geometry of the measuring chamber is shown in Figure 3-7, and the experimental set-up is shown in Figure 3-1. The approximate dimensions of the samples are: centerhole diameter 38 mm, ring diameter 90 mm and ring thickness 32 mm. In the He-gas method it is essential that there is no pressure difference between the end faces of the sample so that gas migration takes place by diffusion only.
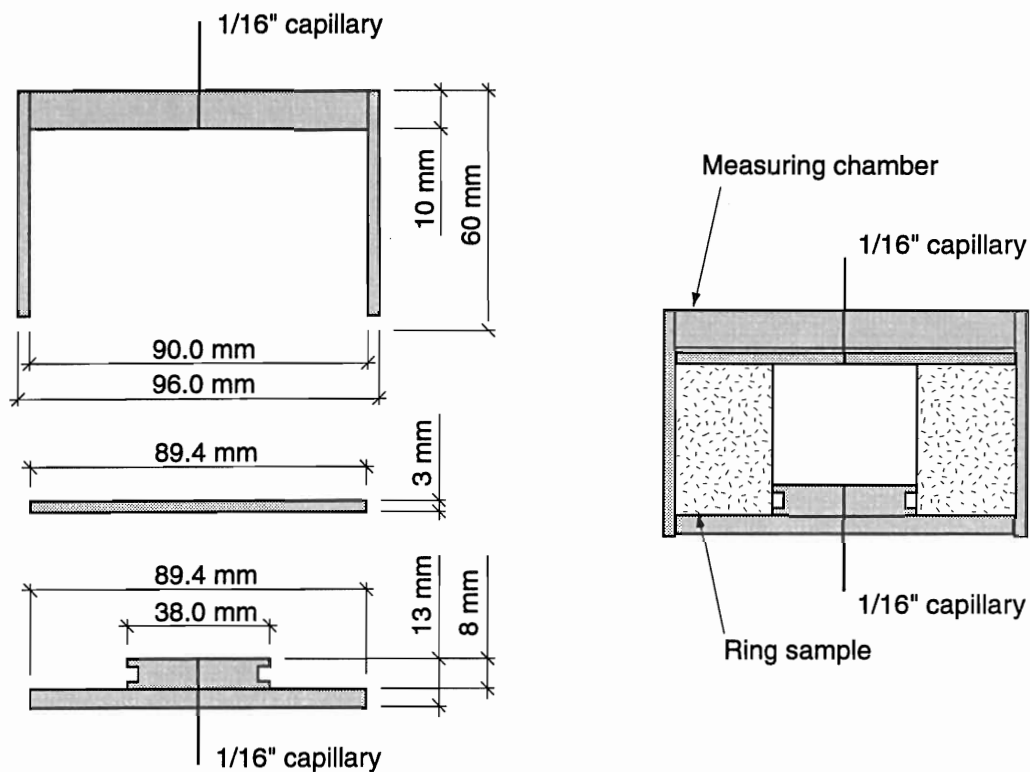
Figure 3-7. Measuring chamber for ring samples.

3.3.1.2   Cubic samples

The principle of measurement for cubic samples is similar to that for ring samples. In the case of cubic samples the helium pulse is injected into the injection chamber so that one rectangular face of the sample is exposed to a known quantity of helium. The opposite face of the sample is continuously flushed with nitrogen while the helium is diffusing through the sample in order to catch the helium molecules which have diffused through the sample. The helium concentration of the nitrogen is then measured. Samples are kept at ambient temperature and pressure. The experimental set-up for cubic samples is shown in Figure 3-2 and some details of the measuring chamber are shown in Figure 3-8.
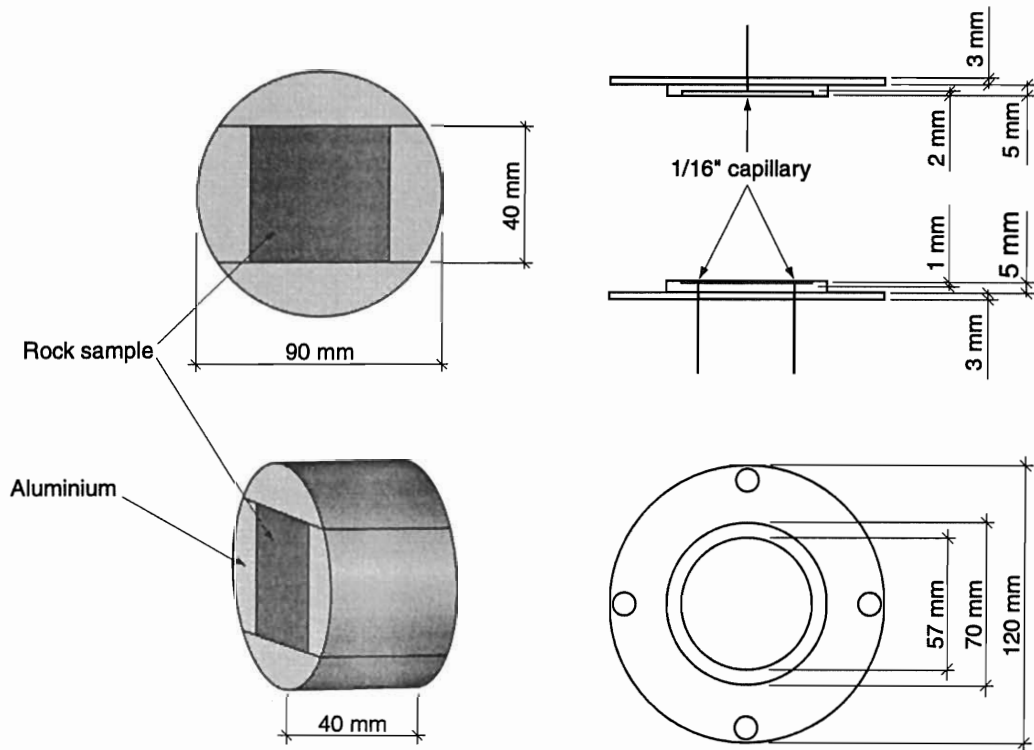
Figure 3-8. *The aluminium parts that make the cubic sample into a cylinder and the end plates of the measuring chamber.*

3.3.2 Modelling and interpretation

Modelling of the through-diffusion measurements is based on the solution of an appropriate diffusion equation. Because of the planar geometry of the samples and the reflective boundary conditions in the transverse directions, one can use a one-dimensional approximation, and the diffusion equation for the concentration of helium $C(x,t)$ is simply

$$\frac{\partial C}{\partial t} = \frac{D_e}{\varepsilon_p} \frac{\partial^2 C}{\partial x^2} \tag{3.3-1}$$

where ε_p [%] is the porosity; C [kg/m^3] is the concentration of helium; x is the distance [m]; D_e [m^2/s] is the effective diffusion coefficient and t [s] is time.

The boundary conditions in the direction of helium transport are such that the concentration of helium in the injection chamber is assumed to decrease according to the diffused amount. It is also assumed that there is no sorption and that the helium concentration at the outer surface of the sample is zero due to continuous flushing with nitrogen.

Porosity ε_p [%] and the effective diffusion coefficient D_e [m^2/s] can both be determined from the breakthrough curves of helium by fitting the curves with an appropriate solution of Equation 3.3-1. In this one-dimensional approximation it is in fact possible to derive (Väätäinen 1994, Carslaw 1959) an analytical expression for the mass flow of helium through the sample,

$$\dot{m} = -D_e A \frac{\partial C}{\partial x}\bigg|_{x=1} \quad [kg/s] \tag{3.3-2}$$

in the form

$$\frac{\dot{m}}{m_0} = D_a h \sum_{n=1}^{\infty} \frac{2h\alpha_n e^{-D_a \alpha_n^2 t}}{\sin(\alpha_n l)\left[l(\alpha_n^2 + h^2) + h\right]} \tag{3.3-3}$$

where A[m^2] is the cross-sectional area of the sample; m_0 [kg] is the mass of the injected helium; $D_a = D_e/\varepsilon_p$ [m^2/s] is the apparent diffusion coefficient; $h = \varepsilon_p/s$ with s [m] being the average length of the injection cell; α_n, n = 1,2,..., are the roots of $\alpha\tan(\alpha l) = h$ [1/m]; 1 [m] is the average length of the sample; and t [s] is time. The average length of the injection cell (s) and the length of the sample (l) are both needed as input parameters.

Once the above mentioned parameters have been fixed, the theoretical model is fitted to the breakthrough curves by varying the effective diffusion coefficient D_e and the porosity ε_p. Another possibility is to infer the value of porosity from independent measurements, in which case the effective diffusion coefficient is the only parameter needed.

Although measurements are carried out in the gas phase, the corresponding diffusion coefficients for the water-saturated case can be derived. This is based on the fact that in both cases, the diffusion equation is the same apart from the actual values of the diffusion coefficients.

For helium diffusing in free nitrogen gas at 22°C, the diffusion coefficient $D_e(N_2)$ is $6.75*10^{-5}$ m^2/s, and for helium diffusing in free water at 22°C the diffusion coefficient $D_e(H_2O)$ is $5.8*10^{-9}$ m^2/s. The ratio $D_e(H_2O) / D_e(N_2)$ is therefore 1/11 600. Scaling the measured diffusion coefficients by this factor should provide a good estimate of the corresponding coefficients for helium atoms diffusing in rock samples saturated with water. If other inert molecules are considered, their mass and size should also be taken into account. For molecules heavier than the helium atom, the diffusion coefficient in free water is typically close to $2*10^{-9}$ m^2/s. Measurements can therefore be scaled to the diffusion coefficients of these heavier molecules in water-saturated samples by using an approximate scaling factor of 1/35 000.

3.4 PERMEABILITY MEASUREMENTS

3.4.1 Measuring technique

The experimental set-up for permeability measurements is similar to that used for through-diffusion measurements.

The surface of the sample in the injection cell is exposed to an elevated pressure of helium. This is the inner surface of the central hole in the case of the ring samples and one of the rectangular faces in the case of the cubic samples. A constant pressure difference is maintained between the surfaces which are used for measurement. The surface of the sample out of which the helium flows is flushed with nitrogen and the concentration of helium in the nitrogen is measured. The flow rate of helium through the sample is measured for several pressure differences. For each pressure difference the flow rate is let to stabilise before reading the value.

3.4.2 Interpretation

In the case when a pressure difference exists across the sample, helium transport through the sample takes place by diffusion and also by flow induced by the pressure gradient. According to Fick's and d'Arcy's laws for a compressible fluid, the volume flow of helium through the sample is given by (Dullien 1979)

$$Q = Q_{diff} + \frac{KA}{\mu l} \frac{\left(p_2^2 - p_1^2\right)}{2 p_1} \tag{3.4-1}$$

where Q [m^3/s] is the flow rate through the sample; Q_{diff} [m^3/s] is the flow rate due to diffusion only; K [m^2] is the permeability coefficient; A [m^2] is the cross-sectional area of the sample; μ [Pa s] is the dynamic viscosity of helium gas; l [m] is the length of the sample; p_2 [Pa] is the pressure of helium in the injection cell; and p_1 [Pa] is the pressure of helium at the outlet. The total flow rate through the sample is measured for several pressure differences, and the permeability coefficient is determined from a fit to the measured points using Equation 3.4-1.

The measured permeabilities, K [m^2], can be used to determine the hydraulic conductivities, K_H [m/s], of the samples. These are given by (Dullien 1979)

$$K_H = \frac{\rho g}{\mu} K \tag{3.4-2}$$

where ρ is the density of water (ρ = 997.08 kg/m^3 at 25°C (Kell 1967)); g is the acceleration of gravity at sea level (g = 9.81 m/s^2); and μ is the dynamic viscosity of water (μ = 8.9042*10^{-4} kg/ms at 25°C (Weast 1974 - 75)).

3.5   POROSITY MEASUREMENTS

3.5.1   General

Porosity measurements are carried out using the He-gas pycnometer method which is based on the ideal-gas equation of state. Helium gas is used because it is non-reactive and penetrates into small pores, and because it very accurately fulfils the ideal gas law. Also, the value obtained by using helium gas will obviously be closer to the effective porosity measured in the He-gas diffusion experiment than a value obtained by using some other media such as water. Therefore the values obtained by using He-gas pycnometry are complementary to the interpretation of the diffusion measurements, as the later are quite insensitive to porosity which is included in the Equation 3.3-3 in parameters D_a and h.

Before any measurements are made, samples are dried in a vacuum oven. After drying, grain volumes are measured using a helium gas pycnometer developed by the Department of Physics at the University of Jyväskylä. Bulk volume determinations are based on the water-immersion method. After the first porosity measurements, samples are cut into two halves. One half, which represents undisturbed rock, is used as a reference sample for the second half which includes the disturbed zone.

3.5.2   Drying of the samples

All samples are dried in a vacuum oven at a temperature of 45°C. This slightly elevated temperature is not expected to cause any damage to the rock structure. Before drying, samples are weighed with a balance in ambient conditions. In order to follow the drying process, samples are weighed after suitable intervals. Drying is interrupted during weighing, the oven is filled with nitrogen and the sample is left in the nitrogen atmosphere for four hours. After weighing, drying is immediately continued. The extent of drying is estimated from the weight measurements by using an exponential fit to the drying curve.

The drying of rock samples takes a long time. The samples from the Research Tunnel at Olkiluoto (of size 4x4x8 cm) e.g. were dried for about 2500 hours. Drying time can be significantly reduced by increasing the temperature and it is suggested that the drying temperature be increased to 105°C if significant deformation is not expected to occur because of heating.

3.5.3 Measuring technique

The experimental arrangement of the pycnometer used in the porosity measurements is shown in Figure 3-9. The volumes of the cells, V_R and V_S, are first determined by using calibration samples. A calibration sample of volume V_N is inserted into the sample cell V_S, the whole system is evacuated and the pressure P_V and the temperature T_V are measured after stabilisation. The reference cell is then pressurised with helium. The pressure V_R and the temperature T_R are measured after stabilisation and the helium is allowed to expand into the measuring chamber. After stabilisation, the resulting pressure P_S and temperature T_S of the whole volume are measured. The unknown volumes in the system can be determined when the volume V_N, pressures P_V, P_R and P_S, and the temperatures T_V, T_R and T_S are known for both sets of measurements. The same procedure is then repeated on the sample to be measured.

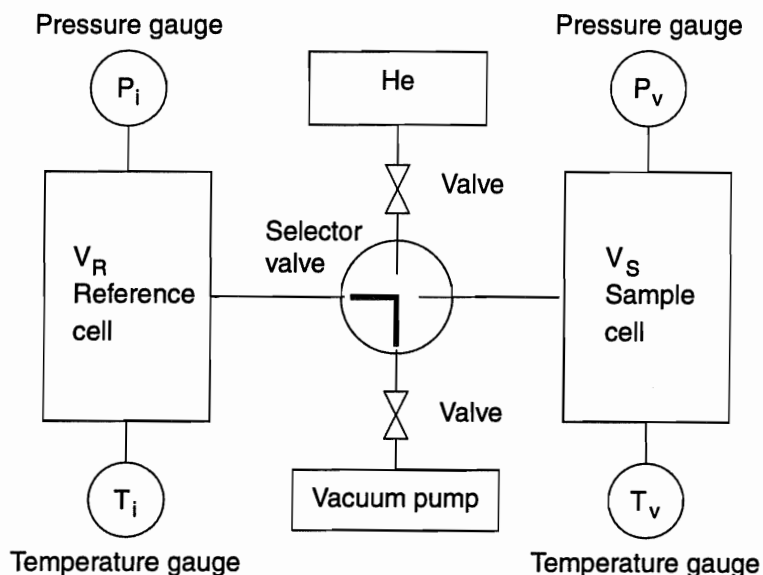***Figure 3-9.*** *Experimental set-up of the He-pycnometer.*

3.5.4 Interpretation

According to the ideal gas law, the porosity of the sample can be determined when the pressures P_V, P_R and P_S, the temperatures T_V, T_R and T_S, the volumes of the cells V_R and V_S, and the bulk volume of the sample V_b are all known. The porosity ε_p [%] can be expressed in the form

$$\varepsilon_p = \frac{V_b - V_g}{V_b} \cdot 100 \qquad (3.5\text{-}1)$$

$$V_g = V_S - V_R \frac{(P_R T_S T_V - P_S T_V T_R)}{(P_S T_V T_R - P_V T_S T_R)} \qquad (3.5\text{-}2)$$

Bulk volume was measured using the water immersion method. The sample was weighed in ambient conditions and then in water. The temperature of the water was also measured to determine its density. The density of water as a function of temperature is given by

$$\rho_w = 0{,}99995 + 0{,}00005 \cdot T - 7{,}545 \cdot 10^{-6} \cdot T^2 + 3{,}6131 \cdot 10^{-8} \cdot T^3 \qquad (3.5\text{-}3)$$

where ρ_w [kg/m^3] is the density of water and T [°C] is the temperature of water. Using the difference in the two masses and the density of water, the bulk volume of the sample can be determined from

$$V_b = \frac{m_d - m_f}{\rho_w} \qquad (3.5\text{-}4)$$

where m_d [kg] is the mass of the sample in ambient conditions and m_f [kg] is the mass of the sample in water.

The mass of the sample at the moment of immersion was determined by using a polynomial curve fitted to the measured points because the mass of the sample grows rapidly during the measurement as water penetrates the pores of the sample.

4 OTHER SUPPORTING METHODS

The ^{14}C-PMMA method is a practical and efficient tool for studying rock structure and the spatial distribution of porosity. Porosity values obtained using the method are close to those resulting from the use of other techniques (Rasilainen et al. 1996) although method-specific differences have been identified. Since the porosities obtained using the ^{14}C-PMMA method are usually compared against porosities measured by other techniques, it is proposed that independent reference measurements are made using other techniques to establish the porosity of both disturbed and undisturbed rock samples. The values thus obtained can then be used to scale any results obtained using the ^{14}C-PMMA method to correspond to other specific methods.

If a description of the different types of microfractures and their location in different minerals is required, it is suggested that investigations be carried out using scanning electron microscopy (SEM) and/or fluorescent microscopy. These techniques will make it possible to obtain a qualitative determination of the distribution of different types of fracturing in different minerals.

It is suggested that fluorescent microscopy is used to verify the degree of penetration of the epoxy-based sealant into the disturbed zone for all the rock types which are of interest.

Different image analysis techniques can be used to support the methods described in this report. They can be used efficiently in:

- studying the distribution of porosity in different minerals,
- accurately determining the position of the disturbed surface of the rock sample, which in turn improves determination of the depth of the disturbed surface, and
- characterising the spatial distribution of disturbance which is seen as increased and deformed microfracturing but which does not significantly affect the porosity of the rock.

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