SEDIMENTOLOGICAL AND PETROPHYSICAL CHARACTERISATION OF THE LOCHALINE SANDSTONE, UPPER CRETACEOUS, NW SCOTLAND

- DETERMINATION OF MICROSTRUCTURAL PARAMETERS IN A PURE QUARTZ SANDSTONE -

By
Gregor Bächle

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Department of Geology
University of Tübingen
(Laboratory work carried out at the University of Aberdeen)
Abstract

Two major periods of marine transgression are recorded in NW-Scotland. The Lochaline sandstone represents a deposition during a minor transgressive phase of Cenomanian age which is interrupting the first trangressive phase. The Lochaline sandstone crops out along a small north-south striking band ranging from Morvern in the north to Mull in the south. Sedimentological analysis of five lateral discontinuous outcrops at Beinn na h-Uamha in Morvern (previously unpublished) yield to a subdivision of the sandstone into four distinctive units. A near-shoreline depositional environment was concluded from the analysed sedimentological facies, the rare ichnological findings and the mineralogy. The range of recorded sedimentary facies may provide useful analogue information for extrapolation into the subsurface, in particular with regard to the hydrocarbon potential of offshore Cretaceous sections.

The pore structure of 22 samples of the Lochaline Sandstone was investigated, employing thin section analysis combined with SEM-image analysis, mercury intrusion, helium porosimetry and complex electrical conductivity measurements, from cores with porosity ranging continuously from 4 - 21%. This large variation was achieved without any change in composition: The Lochaline Sandstone is composed of homogeneous grain size (~240 µm). The low porosity samples were taken from cemented horizons within the formation. Examining thin sections and outcrops revealed that the cemented horizons are caused either by pre-Tertiary surface or subsurface (groundwater) silification, they are genetically not related to the overlying Tertiary basalts.

The apparently complete “pore fraction distribution” relevant for transport was determined using a combination of data from SEM-image analysis and mercury intrusion which enables to measure pores from 0.01 micrometers up to several millimetres in diameter.

Transport properties were studied measuring Klinkenberg corrected gas- and brine permeabilities at 400 psi confining pressure and electrical conductivities at 1kHz.

The results showed an excellent correlation between permeability and measured porosity for all samples. There existed two regimes with distinctly different poroperm relationships: for low helium porosities (\( \phi = 4\% - 12\% \)), permeability (in mD) was \( 7 \times 10^{-4} \phi^{5.59} \), while for high helium porosities (\( \phi = 12\% - 21\% \)), permeability (in mD) was given by \( 12.78 \phi^{1.66} \). The correlation was also excellent between electrical conductivity and porosity. Normalised conductivity (1 / FF) changed with porosity reduction following a single power law (1 / FF = \( \phi^{a} \)) with an exponent of \( a = 1.88 \). The clear correlation between hydraulic and electrical properties with porosity was related to constant grain size, while the two regimes for the hydraulic conductivity emphasised the importance of the microstructure for this process.

“Effective” pore structure parameters like the pore radius related to the capillary threshold or to the entry pressure were derived and used to predict permeabilities and to verify models in permeability prediction in a pure sandstone. Models dealing with characteristic pore length (Pittman, 1992, Katz and Thompson, 1987) underestimate permeability, while the permeability in low permeable samples (K < 10 mD) is overestimated. Predicted permeabilities from models incorporating the grain size (Revil et al., 1997; Van Baaren, 1979; Berg 1970) correlated very well with measured permeability in the low permeable samples, but overestimated permeability as well.

Different techniques in porosity determination were examined, too. The core plug porosity was measured most accurately by helium porosimetry as proved by the grain density, which was closest to the density of quartz. Fluid imbition methods (calliper, Archimedes) slightly underestimated the helium porosity. Porosities measured by mercury injection showed a weak correlation with the core helium porosity. As well, the two-dimensional porosities derived from SEM image analysis and light microscopy point counting were less reliable and tend to underestimate, respectively overestimate in the latter case, the helium porosity.

From the petrophysical point of view, it is established that the Lochaline sandstone has excellent reservoir characteristics.

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**Declaration**

I am assure that I made this thesis on my own and without any unauthorised support.
Nomenclature

- **Hg**: Mercury porosity
- **He**: Helium porosity
- **Arch**: Archimedes porosity
- **Cal**: Calliper porosity
- **PC**: Point counting porosity
- **SEM**: SEM porosity

- **λ**: Wavelength
- **τ**: Electrical tortuosity
- **D**: Fractal dimension
- **d**: Dominant grain diameter
- **FF**: Formation Factor
- **r**: Pore capillary radius
- **G**: Pore geometrical factor
- **p**: Pressure
- **α**: Phase angle
- **q**: Cole-Cole exponent
- **σ_a**: Surface conductivity
- **APore**: Area of pore space
- **Acement**: Area of cement
- **Pc**: Mercury / air capillary pressure
- **Pd**: Mercury / air extrapolated displacement pressure
- **SHg, SRV**: Specific surface (surface area per rock volume)
- **SPV**: Specific surface (surface area per pore volume)
- **SGV**: Specific surface (surface area per grain volume)
- **SFrac**: Fractional mercury saturation
- **SPerc**: Percentage of mercury saturation [%]

- **ρ''**: Imaginary part of the complex resistivity
- **ρ'**: Real part of the complex resistivity
- **σ''**: Imaginary part of the complex conductivity
- **σ'**: Real part of the complex conductivity
- **κ''**: Imaginary part of the complex relative dielectric permittivity
- **κ'**: Real part of the complex relative dielectric permittivity
- **κ_0**: Permittivity in vacuum = 8.85419*10^-12
- **K_{Apex}**: Gas permeability, Klinkenberg corrected
- **K**: Gas permeability, Klinkenberg corrected
- **K_{Apex}**: Permeability predicted by empirical equation after Pittman
- **K_{KC}**: Permeability predicted by Kozeny-Carman model
- **K_B**: Brine permeability
- **K_P**: Permeability measured by probe permeameter
- **R_{Apex}**: Characteristic pore radius calculated after Pittman
- **R_{Thres}**: Characteristic pore radius calculated after Katz and Thompson
- **R**: Hydraulic radius (equivalent to R_{thres})
- **R_{Av}**: Average pore diameter of the pore size distribution derived from mercury injection
- **R_{SEM75}**: Pore radius at the third quartile (75%) of the SEM pore size distribution curve
- **R_{35}**: Pore radius at 35% mercury saturation after Winland
- **R_{SEM50}**: Mean SEM pore radius
- **l_1,Hg**: lower length (radius) limit of the fractal regime calculated from mercury injection
- **l_2,Hg**: upper length (radius) limit of the fractal regime calculated from mercury injection
- **l_1,SEM**: lower length (radius) limit of the fractal regime calculated from image analyses
- **l_2,SEM**: upper length (radius) limit of the fractal regime calculated from image analyses
- **d_{Hg}**: Average grain diameter derived from mercury injection
- **((SPerc)Pc/Pc)_{max}**: Maximal ratio of percentage mercury saturation divided by capillary pressure

- **A_{detrital grain}**: Area of detrital grain
- **A_{intergranular}**: Intergranular area
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I. Introduction

Previous literature of the Lochaline Sandstone (LSst) has concentrated on the sedimentology (Lowden et al, 1992), on the palaeoenvironment (Bailey, 1924; Braley, 1990) and on the mineralogy (Humphries, 1961), rather than on the petrophysical studies (Lewis et al. 1990). In this case study the centre of investigation focused on the petrophysical characterisation of the sandstone. This is done by examination of only few samples by routine as well as by special core analysis techniques. Additionally this pure quartz sandstone with its relative simple pore structure was used to determine pore structural parameters and to verify models dealing with permeability prediction.

Such a study is important for the following reasons:

- The improved characterisation of sandstones increases the petrophysical control we have on data from borehole measurements.
- Sandstones such as the LSst are subsequently used as a standard test rocks for the laboratory examination of various physical mechanisms operating in rocks.

In this context the sedimentological characterisation is only of marginal interest. It is reduced to aspects which could directly influence the petrophysical behaviour of the sandstone, e.g. grain size distribution at micro scale or the genesis of cemented horizons at reservoir scale.

In order to introduce the Lochaline Sandstone an overview of essential previous work is presented in this chapter.

It is also given a short summary over models in permeability prediction over the last few decades to clear the (sometimes similar) parameters.

1.1 Objective

The principle aim of this report is to understand the rock in its fullest detail and to identify its porosity/permeability relationship.

A further subject is to relate predicted permeabilities derived from capillary pressure, porosity and conductivity data to measured Klinkenberg permeabilities.

An attempt is also made to characterise the influence of cement evolution on petrophysical properties and to examine the origin of the cemented horizons.
1.2 Exposure and Stratigraphy

The Lochaline Sandstone Member is part of the Cretaceous Inner Hebrides Group (Lewis et al., 1990). Sediments of Cretaceous age in Northwest Scotland outcrop in small, often isolated exposures throughout the Inner Hebrides and Morvern. The best known and most extensive outcrop is located near Lochaline (fig.2), in and around the Lochaline sand mine. The thickness of the formation varies from approximately 12 metres (Lowden et al., 1992) to occasionally only 5 metres (Braley, 1990) at outcrops around Lochaline whereas the explored sequence within the mine has a thickness of 3 to 8 metres. A borehole drilled 2 km north of the mine entrance reveals a thickness of only 1.46 metres. Similar sequences are also preserved nearby on Beinn na h-Uamha and on Beinn Iadain and on Western Mull (Judd, 1878; Bailey, 1924; Lee and Pringle, 1932) which are presumed to be the lateral equivalent of the Lochaline Sandstone (Lewis et al., 1990).

The Lochaline Sandstone occupies an intermediate position between a glauconitic Greensand and a red mudstone on the west side of the Loch Aline. Since only one fossil, a starfish (McLennan, 1949), has been found in the sandstone, its biostratigraphic status remains uncertain. However, the sequence is traditionally interpreted to be of Cenomanian age based on Cenomanian fossils (Judd, 1887) from the underlying Greensand and on a thin clayey band reach of fossils of Cenomanian age at the top of the Lochaline Sandstone at Beinn Iadain (Lee et al., 1932). The age of the Greensand was recently confirmed by microfossils (Lewis et al., 1990).

During the Cenomanian two periods of marine transgression in the Inner Hebrides basin occurred. The first began in the latest Albian and continued through the early Cenomanian, as represented by the deposition of the marginal clastic facies of the Morvern Formation (fig.1). The Lochaline Sandstone Member is thought to be a deposited in a minor period of regression in the late Cenomanian (Braley, 1990).

Fig.1: Lithostratigraphy for the Cretaceous of NW-Scotland (after Lowden et al., 1992).
Fig. 2: Examined locations of outcrops of the Lochaline Sandstone at Loch Aline and at Beinn na h-Uamha.
1. 2 Review: Sedimentology of the Lochaline Sandstone

1. 2. 1 Mineralogy and Grain Size
Previous examinations were carried out exclusively on exposures within and around the mine. The sand consist almost entirely of the mineral quartz (~99 % by weight).
Sieve analysis have approximately log-normal distribution, but grains below 0,19 mm deviated from this significantly (Humphries, 1961). The arithmetic mean grain sizes are ranging from 0.177 mm to 0.244 mm (Lewis et al, 1990) or 0.238 mm accordingly to Humphries, 1961.
In most of the samples 70% of the sand is between 0.2 and 0.3 mm, 10% is coarser than 0.3 mm and only 3% is finer than 0.08 mm (Humphries, 1961). The arithmetic mean size is positively related to the mean grain roundness by a power function. For grains smaller than 0.2 mm the roundness falls off much more rapidly with decreasing size. Lewis et al. (1990) noticed a large negative skew in the fine tail of the grain size distribution frequency histograms which may represent a second mode in the finer fractions with its own distribution characteristics. This would be in agreement with Humphries observations.
Lewis et al. (1990) recorded two vertical coarsening upwards sequence in which silty sands of variable thickness (up to 3 metres) pass upwards into medium-grained, well sorted sands. Lowden et al. (1992) noted that at some exposures the coarsening upwards transition is as short as less than one metre of the overall sequence.
At one locality, in the roof of the mine, exists an exposure of medium to large, subrounded to rounded, pebble-sized quartzose clasts accordingly to Lewis et al. (1990). Braley (1990) observed also an increase in grain size in the top 40 cm at an outcrop 1.5 km northwards on the western side of the Loch Aline. Pebbles were recorded which are up to 1.2 cm long.
An idealised vertical sequence through the unit is presented in Figure 3. It shows a general fine to medium sand coarsening-upwards sequence.

1. 2. 2 Sedimentary and Biogenic Structures
In general there are only few sedimentary features visible due to the nature of the well sorted pure quartz sand, but can be observed to a slightly greater extend on weathered surfaces outside the mine. Cross-bedding structures are occasionally highlighted by iron-precipitation. Lewis et. al. (1990) noted bioturbation of Ophiomorpa sp. and Skolithos sp. restricted to silty horizons at the base of the Lochaline Sandstone. Lowden et. al. (1992) recognised also that bioturbation is particular abundant near the sharp basal contact with the underlying Greensand. Subvertical to horizontal burrows of the Thalassinoides-type ichno assemblage are overprinted by thin vertical burrows of the Skolithos-type at outcrops around the mine (Braley, 1990). The lateral extension is limited to few metres and also the degree of bioturbation varies significantly.
Above the bioturbated unit is a scoured interval containing ripples, rare swaley / hummocky
cross stratification and some soft sediment deformation. The upper part shows isolated Ophimorpha burrows and rare cross beds of approximately 30-40 cm thickness, current ripples, slump structures and apparently bundled silty laminations (Lowden et. al., 1992 and Lewis et. al., 1990). The lack of sedimentary structures was attributed to storm reworking events. The wavelength of slightly asymmetric ripples appears to be an average of 40 cm, the ripple set thickness averages 18 cm (Lowden et. al., 1990). The cross-bed units are suggested to have a lateral extension of at least 100 m, since a number of units can be traced over tens of metres with remaining constant thickness and dip. Foreset orientations recorded a unidirectional north-western current flow.

In the midst of the Lochaline sandstone at Beinn na h-Uamha (Beinn-y-Hun) a thin seam of lignite or coal has been found (Judd, 1878). Braley (1990) also noted that following a erosive contact fine grained sands containing limonite horizons are in places concentrated into parallel laminae and also containing horizontal burrows. This horizon grades up into a homogeneous fine grained sandstone, representing the Lochaline Sandstone. Hard bands, approximately 0.5 cm thick, are developed towards the bottom and top of the Lochaline sandstone in and around the mine.

Fig. 3: Vertical sequence through the Lochaline sandstone, without hard bands (Lowden et al., 1992).

1. 2. 3 Depositional Environment
Various depositional environments have been suggested for the Lochaline sandstone. But nevertheless all interpretations (Bailey, 1924; Mac Lennan, 1949; Humphries, 1961; Braley, 1990; Lewis et. al., 1990; Lowden et al., 1992) are in agreement with a shallow marine environment.

Most of the investigations were carried out around or within the easy to reach Lochaline sand
mine. Investigations by the Geological Survey of Scotland (Braley, 1924) came to the hypothesis that the LSst represented a desert sandstone finally blown into the sea, based on perfectly rounded grains. Humphries (1961) argued from his petrological point of view towards a reinterpretation of the unit as a high energy, shallow marine water facies of Greensand where constant reworking resulted in removal of the soluble and less stable constituents. Lewis et al. (1990) noticed within the mine a bioturbated base and interpreted it as a lower shoreface deposit, the upwards decrease in bioturbation as an upper shoreface deposit. The lack in sedimentation structures was explained by storm reworking activity. Lewis et al. (1990) also suggested that the sandstone may represent the reworking of the underlying Greensand by tidal and/or wave action. Lowden et al. (1992) noted a coarsening upward sequence and interpreted it together with the regional setting as a wave dominated, prograding shoreface. The fine and highly bioturbated base was seen as a offshore-transition zone.

Only few research were carried out on the more remote interior outcrops of the Lochaline sandstone: Judd (1887) interpreted the LSst as an estuarine deposit, based upon the apparent presence of much carbonaceous matter and a seam of coal at Beinn Iadain, as well as on the relations to the “marine series”, i.e. the underlying Greensand. Braley (1990) examined the sandstone at several localities and noted a distribution of outcrops which appears to be parallel to the trend of the Great Glen Fault. Nevertheless the distribution of outcrops should not be overrated since there is only a limited exposure of the LSst. Braley suggested that the LSst represents a bar or shoal deposited in following environment: wave direction across the Inner Hebrides Basin from north west meeting fluvial currents inferred down the lineament of the Great Glen Fault.

1.2.4 Diagenesis

Previous research revealed that the sandstone is in general terms little altered. The diagenetic features are only qualitatively described so far and include:

- compaction (Lewis et al, 1990 and Lowden et al., 1992)
- decalcification (Lewis et al, 1990 and Lowden et al., 1992)
- iron staining (Braley, 1990 and Lewis et al., 1990)
- secondary silification (Humphries, 1961; Braley, 1925; Mac Lennan, 1949; Lewis et al., 1990 and Lowden et al., 1992)
- contact metamorphosis due to dykes (Lewis et al., 1990; Lowden et al., 1992 and Munro, 1997), but cementation and iron staining only in a distance of less than a metre.

Braley (1925) first recognised the frosted surfaces of many grains in the hard bands which Mac Lennan (1949) interpreted later as evidence for Aeolian abrasion. The frosting, which is restricted to the hard bands, is evenly spread over the surface of grains of all sizes and it is also present in pits, grooves and fractured surfaces. Thus Humphries (1961) has been
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questioning mechanical abrasion under Aeolian conditions to have caused frosting. He also noted a variable degree of facets from small quartz facets of minute ridges up to completely envelopment of the grain. Lewis et al. (1990) concluded that frosting occurs as a diagenetic process and that the quartz overgrowth has produced the faceting. A sometimes visible interface between quartz grain and secondary quartz layer was suggested to be a attribute to the presence of some opaque coating on the original grain, or a layer of inclusions within the grain or the secondary silica (Humphries, 1961). He assumed that it is “likely that solution of fine quartz particles has contributed to the supply of silica” because he noted that the development of facets is more pronounced on the coarser grains, suggesting that the precipitation of silica is connected to the curvations of the grains and so with its surface energy. The formation of distinctive hard ribs explained Humphries by the culmination of the silification process in that horizons.

The silification has been associated with the silification of the overlying chalks at Beinn Iadain (Braley, 1924; Mac Lennan, 1949). It was suggested from Lowden et al. (1992) that silification occurs during Cenomanian since almost exclusively clasts of silicified chalk (and flint clasts) were found in paraconglomerates of the Turonian Laig Gorge Sandstone Member (Braley, 1990). At two localities Braley recorded that the facies lies unconformable on the Morvern Greensand, but no reworked cemented sandstone were found.

Lewis et al. (1990) described a hard persistent quartz band forming the roof of the mine and a second discontinuous wavy and often lens shaped quartz band which joints up at some exposures from the overlying rib. The bands are approximately 0.5 metres thick (Lowden et al., 1992). The roof band seems to be unrelated to any depositional structures, nevertheless for the hard bands primary factors such as highly permeable coarse grained cross-bedding units (Lewis et al., 1990) and possible concentrations of biogenetic silica (Lowden et al., 1992) were considered. Since iron staining is mainly concentrated in the 2 to 3 metres above the lower hard band, Lewis et al. (1990) suggested that this hard band had acts as a barrier for downwards percolating iron rich groundwater. It was also noted that iron oxide sometimes appears to follow sedimentary structures. Braley (1990) also noted that iron rich horizons follow joints and cracks which suggests that it is probably a recent ongoing process. Iron precipitation restricted to faults which could fill the entire original pore volume are examined by Munro (1997).

Lewis et al. (1990) add on a decalcification process due to the absence of calcareous fossils. Fracturing of quartz grains is difficult to assess since the effects of blasting and face preparation are not easily to be distinguishable from natural fracturing. But compaction is best observed at the few micas which appear to be squeezed between the more robust quartz grains. Since Lewis et al. (1990) observed also fractured grains which are displaced and forced into pore space, a not too negligible amount of natural compaction have to be considered.
1.3 Review: Models in Permeability Prediction

As stated above this study is focused on properties which are useful to predict permeability as well as on the characterisation of the pore structure. The latter includes the size of the pores and throats and its three-dimensional constrictions, and will massively influence the permeability of a porous media. It is therefore of advantage to relate the pore structure to petrophysical properties to predict permeability. A large amount of different theoretical models have been developed to account for the ecological as well as economical importance of the ability of permeability prediction. The literature cited below is not comprehensive but gives a rough overview over models employed to predict permeability:

- Equivalent channel (Walsh & Brace, 1968, 1984; Paterson, 1983)
- Effective medium (Koplik et al., 1994; Doyen, 1988)
- Statistical (Gueguen & Denies, 1989; Gueguen, David & Darot, 1986)
- Percolation (Katz & Thompson, 1987; Zhu, David & Wong, 1995; Gueguen, 1996; Mavko & Nur, 1997; Wong, Koplik & Tomanic, 1984))
- Networks (Bryant et. al., 1992 & David et al., 1990)
- Fractal (Adler, 1989; Ahernov, 1997; Hansen & Skjeltop, 1988; Aharonov et al., 1997; Pape et al., 1984)
- Pore scale (Plessis, 1994)
- Kozeny-Carman (Kozeny, 1927; Carman, 1937; Denicol & Jing, 1996)
- Empirical (Swanson, 1981; Pittman, 1992, Kolodzie, 1980)
- Grain size (RGPZ, Berg, 1970)

Some of these models are reviewed and examined for their dependence on porosity by Nelson (1994).

Within this study, three major models were selected and employed for data analysis:

- The Equivalent Channel Model and Modifications (1)
- The Bundle of Tube Model and Modifications (2)
- Grain Size Models (3)

They are the most common models and the parameters can be obtained from mercury injection and/or conductivity measurements.
1. Equivalent Channel Model and Modifications

While grain size has a negligible effect on the porosity of a rock, this parameter has a predominant effect on permeability. The reason for this is that grain size controls the total wetted surface - and shearing friction is formed between the zero layer velocity on the surface and any passing fluid. The earliest attempt to relate permeability to the internal surface area of a rock was performed by Kozeny (1927) and he derived the following relation for consolidated media:

\[ k = \frac{\text{SpV}}{(2 \times \text{SpV}^2)} \]

where
\[
\text{SpV} = \text{surface area per unit pore volume}
\]

Carman (1937) adapted the original Kozeny equation for formations comprised of uniform and homogeneous rocks having a dominance of nearly spherical grains for unconsolidated media:

\[ k = \frac{3}{(c \times \text{SGV}^2 \times (1 - \phi^2))^2} \]

where
\[
\text{SGV} = \text{surface area per unit grain volume}
\]
\[
\phi = \text{porosity [%]}
\]
\[
k = \text{permeability [Darcy]}
\]
\[
c = \text{constant based on experimental data on packed beds ~ 5}
\]

One should note that the specific surface is dependant of the scale and resolution of the technique employed. The reciprocal of the ratio of surface area to pore volume is also known as the "hydraulic radius" R.

Originally the equation corresponds strictly only to through-going tubes of a constant cross-section surface. But as Paterson (1983) clearly reviewed, the Kozeny-Carmen model can also be theoretically obtained by combining the Hagen-Poiseuille law with Darcy’s law:

\[ k = \frac{3}{(a \times \tau \times \text{SRV}^2)} \]  
-> (pore surface area normalised by rock volume)

or

\[ k = a \times R^2 \times \frac{\phi}{\tau} = a \times R^2 \times \phi^m \]  
-> (pore surface area normalised by grain volume)

where
\[
a = \text{numerical factor ~ 400}
\]
\[
\tau = \text{tortuosity} \quad k = \text{permeability [mD]}
\]
\[
R = \text{hydraulic radius [$\mu$m]} \quad \phi = \text{fractional porosity}
\]
This most frequently cited modified Kozeny-Carman equation takes the tortuosity into account and therefore partially replaces the experimental constant. Through introduction of electrical properties the porosity term can be fully eliminated (Paterson, 1983 and Walsh & Brace, 1983).

One should note that one assumption of this model is a lack of channelling in the porous media. The hydraulic radius makes also the implicit assumption that all pores of all sizes will be weighted equally in permeability prediction. It is therefore not surprising that the apparent failure of the K-C relation (porosity power of two) is often observed in the low porosity range where permeability decreases more rapidly with decreasing porosity than in the high porosity range.

More recently published work by Achdou and Avellaneda (1992) examines the empirical formula of JKS which is based on the relation between hydrodynamics and electricity:

\[ k = S_{el}^2 / 8 \text{FF} \]

where

- \( S_{el} \) = JKS length scale, the electrically weighted specific surface
- \( \text{FF} \) = Formation factor

They conclude from computer simulations and simple models that permeability for in-parallel model is an average of single tube responses, and a harmonic average for the in-series model. They show that the JKS equation underestimates permeability in the former case, where the JKS length scale will always be smaller than the effective hydrodynamic radius (e.g. derived from mercury intrusion) and overestimates permeability in the latter. The electrical theory accompany much better the real behaviour in the in-series model because both of them, the electrical and the hydrodynamic effective radius, are dominated by the small pore throats. Significant deviations occur only in the case of a broad pore size distribution at small scales.
2. Bundle of Tube Models and Modifications

The bundle of tube model was established to describe in a simplistic way the complex pore system of a porous media by measurement of the capillary pressure. If the porous media is conceived as a bundle of capillary tubes of various radii, then the capillary pressure-saturation curve relates to the number and size of pores penetrated by the non-wetting fluid at a given capillary pressure. Washburn (1921) first suggested the measurement of pore aperture size distribution by the use of mercury injection. He derived his equation from the Hagen-Poiseuille form of the Laplace equation. It is expressed as:

\[ P_c = -\frac{(2\sigma \cos \theta)}{r} \]  

(see Chapter 3.1.1. for details)

where

\( \sigma \) = surface tension  \( P_c \) = capillary pressure  
\( \theta \) = contact angle  \( r \) = capillary radii

Based on Washburn’s model, Purcell (1949) has developed an empirical relation for estimating permeability from mercury injection capillary pressure data. His equation combines the capillary pressure definition, Poiseuille’s and Darcy’s law and can be expressed as

\[ k = \left(\frac{(\sigma \cos \theta)^2}{2}\right) \Sigma \left( \frac{S_{perc}}{(P_c)^2} \right) \]

where

\( k \) = permeability \( [\text{mD}] \)  \( S_{perc} \) = Percentage of mercury saturation \( [\%] \)  
\( P_c \) = capillary pressure \( [\text{psi}] \)  \( \Sigma \) = porosity

To account for the deviation of the actual pore space from the simple geometry used in the derivation, Purcell introduced a lithology factor “L” into the final equation. After introducing conversion factors and the standard values for mercury intrusion (\( \sigma = 480 \), \( \theta = 140^\circ \)), the equation could be reduced to

\[ k = 14.260 \Sigma L \left(\frac{dS}{P_c^2}\right) \]  

(after Cortez, 1993)

The term \( \left(\frac{dS}{P_c^2}\right) \) represents the distribution of the percentage of contribution of the pore radii for the ability of mercury-flow (Deuterer and Menz, 1984). The prediction of permeability with this formula which relates permeability to an integral of \( P_c^2 \) over the entire saturation range, places greater emphasise on the larger pores than on the smaller pores (which have low capillary pressures).

One decade later, Thomeer (1960) introduced a new analytical technique based on the
location and shape of capillary curves in a log-log plot of pressure versus fractional bulk volume occupied by mercury. He suggested that the total interconnected pore volume (at infinite pressures) and the extrapolated displacement pressure (at zero mercury injected) could probably sufficiently characterise the pore structure of the sample measured sufficient. The approach implies that the smoothed curves should represent true hyperbolas. This hyperbola curves can then expressed by the following equation:

\[
\frac{(S_{\text{Frac}})P_c}{(S_{\text{Frac}})P} = e^{-G/(\log P_c/P_d)}
\]

where
- \((S_{\text{Frac}})P_c\) = fractional mercury saturation at pressure \(P_c\)
- \((S_{\text{Frac}})P\) = fractional mercury saturation at infinite pressure or total interconnected pore volume
- \(P_c\) = mercury/air capillary pressure [psi]
- \(P_d\) = mercury/air extrapolated displacement pressure [psi]
- \(G\) = pore geometrical factor

The shape of the curve is defined by the geometrical factor “G” (fig.4), which takes into account the relation of the shape of the curve to the sorting and interconnection of the pores in the rock sample. Hence, it eliminates the need to independently determine a lithology or pore geometry parameter.

Thomeer noted also that “G” adjusts the basic permeability to \((S_{\text{Frac}})P\) / \(P_d\) relation for differences in pore geometry between rock samples. The original relationship was refined with a larger data set and resulted in the following equation (Thomeer, 1983):

\[
k_{\text{air}} = 3.8068 G^{-1.3334((S_{\text{Frac}})P) / P_d)^2}
\]

where
- \(k_{\text{air}}\) = air permeability [mD]
- \(G\) = pore geometrical factor

In 1977 Swanson proposed a method to determine a pressure point on the mercury injection curve which relates to pore sizes interconnecting a continuous pore system through the rock - visualised by injection of Wood’s metal. This point can be graphically determined in a log-log plot of \((S_{\text{Frac}})P_c\) versus \(P_c\). If the scale values are the same, a line drawn at a angle of 45° slanted to the left can be shifted to find the tangent to the capillary curve. The intersection of the tangent with the capillary curve is the point of maximum ratio of mercury saturation to pressure. Because neither the approach of Purcell nor Thomeer’s parameter are always predicting permeability reliably - which is in particular true for drill cuttings - Swanson (1981) created a relation based on 319 cleaned sandstone and carbonate samples from 74 formations:
\[ K_{\text{air}} = 339 \left( \frac{(S_{\text{perc}})P_c}{P_c} \right)_{\text{max}}^{1.691} \]

where
- \( K_{\text{air}} \) = air permeability [mD] at 1000 psi confining pressure
- \( (S_{\text{perc}})P_c \) = maximum ratio of percentage mercury saturation to capillary pressure

He obtained a similar relationship for brine permeability \( K_b \) of 58 clean sandstone and carbonate samples at 1000 psi confining pressure. The power of the related “effective” pore size in prediction of permeability increases to approximately two:

\[ K_b = 355 \left( \frac{(S_{\text{perc}})P_c}{P_c} \right)_{\text{max}}^{2.005} \]

where
- \( K_b \) = brine permeability [mD]

This equation enables to compare the permeability values directly with measured Klinkenberg corrected gas permeability.

At the same time Schowalter (1979) examined the pressure required to form a connecting path of fluid through the largest connected pore apertures of rock samples. Since at low injection pressures the mercury intruded is difficult to determine due to rough surfaces and closure effects, he measured the electrical conductivity simultaneous to injection. A first conducting path was detected at mercury saturations ranging between 4.5-17%. Because on existing mercury data no conductivity measurement were carried out, he defined for his purpose the term displacement pressure as the pressure at 10% mercury saturation.

Winland introduced at first a relation between permeability, pressure from capillary pressure curves and porosity published by Kolodzie, 1980. His data is based on 322 samples, including 82 samples from which Klinkenberg corrected permeability values were available. Under employment of Washburn’s equation he has converted the pressure data into pore aperture sizes. He has carried out regression analysis with the radii as a dependent variant which resulted in the best correlation at a mercury saturation of 35%:

\[ \log R_{35} = 0.732 + 0.588 \log K_{\text{air}} - 0.864 \log \phi \]

where
- \( \log R_{35} \) = pore radii at a mercury saturation of 35%
- \( K_{\text{air}} \) = air permeability [mD]
- \( \phi \) = porosity [%]

Katz and Thompson (1987) reported the following relationship based on data of 60 sandstone and carbonate samples (in parts Klinkenberg corrected) which allows to incorporate percolation theory with parameter gained from the capillary pressure curve:
\[ K_{\text{air}} = \frac{((R_{\text{thres}} \times 2)^2 \times \text{FF})}{266} \times 1000 \]

where
- FF = formation factor
- \( R_{\text{thres}} \) = threshold radius [µm]
- \( k_{\text{air}} \) = air permeability [mD]

The threshold radius is related to the pressure at the first inflection point of the capillary pressure curve at which the mercury forms a connected pathway through the sample. This was demonstrated by continuously monitoring the electrical resistance across the sample and determining the pressure at which electrical continuity is archived. Compared to the Kozeny-Carmen equation, the only difference lies in the conceptional approach and the constant chosen. Because the constant in the equation of the K-C model is approximately 22 times higher, the associated hydraulic radius must be statistically 4.7 times smaller than the characteristic radius of the Katz and Thompson model in the same sample. Only in samples where the flow channels are all equal and of uniform radii one could assume a close correspondence between the two quantities.

Pittman (1992) introduced a new approach to link Winland’s results to Swanson’s theory. He established empirical equations of the relationship of porosity, uncorrected air permeability and various parameters derived from capillary pressure curves using multiple regression on a database of 202 samples of sandstone from 14 formations. Based on a multiple regression with a defined pore radius as the dependant, the relationship among the pore radii, permeability and porosity yielded to following results:
Radii corresponding to Schowalter’s (1979) displacement pressure and Katz and Thompson’s (1986) threshold pressure revealed the same degree of correlation with porosity and permeability (correlation coefficient of 0.9). Radii corresponding to the apex reveal a relationship with a slightly better correlation coefficient of 0.919. Using permeability as the dependant variant resulted in the following regression equation:

\[ K_{\text{air}} = 0.1377 \times \text{porosity}^{1.185} \times R_{\text{apex}}^{1.627} \quad \text{(correlation coefficient = 0.928)} \]

where
- \( \text{porosity} \) = porosity [%]
- \( R_{\text{apex}} \) = pore radius corresponding to the apex [µm]

The apex was determined graphically in a plot of the ratio of mercury saturation to corresponding capillary pressure versus mercury saturation and is equivalent to but more accurate than the apex of Thomeers’s (1959) hyperbola. The mean apex had a mercury saturation of 36%. One should note that the pore radius related to the apex in Pittman’s model...
is associated with the capillary pressure point of Swanson’s model. Since Winland’s equation yields also the highest correlation coefficient at 35% mercury saturation, it is thought that statistically both the semi-theoretically model of Swanson and the two empirical approaches of Pittman and Winland refer to the same point of the capillary pressure curve. Pittman explains the correlation at a point of 35% mercury saturation because that is where "the pore network is developed to the point of serving as an effective pore system that dominates flow in the sense described by Swanson". A second important conclusion was that the porosity term is only significant for higher percentiles of mercury saturation (>40%). Nelson (1994) explained this fact by means of the information that the radius contains regarding the large through-going pores. The higher the content of information containing the radii the lower the dependence on porosity.

Kamath (1992) examined the accuracy of predicting Klinkenberg air permeability from published models dealing with different length scale parameters from mercury injection curves. He concluded from statistical tests (bootstrapping) of data from 66 sandstone and 36 carbonate samples (Chevron data base) that the length associated with Swanson’s maximum value of $\text{Sat}_{\text{Hg}} / \text{Pc}$ will have a better predictive ability than the length related to the entry pressure. No difference appears to exist between the ability of prediction of Swanson’s length scale and the length scale referring to Purcel’s and to Katz and Thompson’s model. This fact is somehow inconsistent, since the Katz and Thompson length scale equals the length scale related to the pore entry pressure.

He developed also a "new" equation based on a data base of 201 sandstone and 99 carbonate samples encompassing a permeability range from 1 mD to 2000 mD. He used Swanson’s length scale for correlation with Klinkenberg corrected air permeability values which yield the following regression equation which is in good agreement with Swanson’s results:

$$K = 347((S_{\text{Perc}})\text{Pc} / \text{Pc})_{\max}^{1.6}$$

where

- $K$ = Klinkenberg corrected air permeability [mD]
- $((S_{\text{Perc}})\text{Pc} / \text{Pc})_{\max}$ = maximum ratio of percentage mercury saturation to capillary pressure [% / psi]

The 95 % prediction interval of the equation is a factor of 5.4, for a limited data set consisting of samples > 10 mD the estimation ability improves and lies within a factor of 3.3. The Katz and Thompson model yielded an underestimation of permeability by an average factor of more than 2. Nevertheless the data of Katz % Thompson’s own data base had the same standard error as the length scale of Swanson applied for the Chevron data base.
The characteristic pore radii used in the previous presented models are summarised in figure 5. An idealised capillary pressure curve for the Lochaline Sandstone is used to show the measures of the different characteristic pore radii. The reciprocal capillary pressure is proportional to the “smallest pore size on the connected path of pores containing largest pores” for the Katz and Thompson model at the entry pressure (point 2). The “pore size connecting the effective pore space” is proportional to the reciprocal capillary pressure at the apex (point 4) for Pittman’s model. The characteristic pore sizes of all models with exception of Purcell’s parameter differ not significantly.

1. Schowalter (1979)
3. Winland (Kolodzie, 1980)
5. Purcell (1949)

Figure 5: Idealised capillary pressure curve showing measures used directly or indirectly by the different authors for determination of characteristic pore dimension.

More recent published work covers the simulation of mercury intrusion and the calculation of absolute permeability in porous media via network models (Matthews et al., 1995) and the modelling of mercury intrusion under incorporation of the percolation theory (Xu et al., 1997).
3. Grain Size Models

*Berg* (1970) published one of the first models which links directly grain size with permeability. From consideration of only rectilicular pores (those pores which penetrate the porous medium without change in shape or direction) of various packings of spheres, he developed an equation, which relates permeability to the square of the grain diameter:

\[ K = 80.8 \cdot 5.1 d^2 e^{-1.385s} \]

where

- \( K \) = permeability [mD]
- \( m \) = cementation factor (= 1.8)
- \( \phi \) = fractional porosity
- \( d \) = median grain diameter [µm]
- \( s \) = sorting term

The sorting term \( s \), also called the percentile derivation \( (s = S_{90} - S_{10}) \), incorporates any spread in grain size into the formula and is expressed in phi units, where \( \phi = -\log_2 d \) (mm). For example, a sample with a median diameter of 0.177 mm, a value of 1 for \( s \) implies that 10 percent of the grains are larger than 0.25 mm and 10 percent are smaller than 0.125 mm. This configuration \( (s=1) \) was chosen to calculate the poroperm trendlines of varying grain diameter in Chapter 4.3.5 and results in the following equation:

\[ K = 20.2 d^2 \cdot 5.1 \]

*Van Baaren* (1979) developed an empirical equation to relate porosity and a dominant grain diameter to permeability, based of the Kozeny-Carmen equation. But instead of using the indirect parameter of the specific surface, he employed directly the dominant grain size:

\[ K = c d^2 \phi^{3.64+m} \]

where

- \( K \) = permeability [mD]
- \( m \) = cementation factor (= 1.8)
- \( \phi \) = fractional porosity
- \( d \) = dominant grain diameter [µm]
- \( c \) = constant related to sorting (25.9-very well sorted)

*Revil et al.* (1987) derived their model (called RGPZ-model) from the consideration of surface conduction theories:

\[ K = (1000 d^2 \phi^{3m}) / (4 * (p) * m^2) \]

where

- \( K \) = permeability [mD]
- \( m \) = cementation factor (= 1.8)
- \( \phi \) = fractional porosity
- \( d \) = dominant grain diameter [µm]
- \( p \) = packing term (for perfect sperical grains = 8/3)

One should note that all three models relate permeability to the square of the grain diameter and to porosity raised to a power of ~ 5.
II. Sampling & Techniques Employed

The techniques employed for examination of the Lochaline Sandstone are X-ray analysis, core analysis and sedimentary logs. The X-ray analysis and the core study are carried out on samples obtained from the LSst mine which is mined in a room-and-pillar principle. The sedimentary logs of the LSst are recorded few kilometres further in the north, at the southern side of Beinn na h-Umaha.

Core sampling was carried out on two already existing unorientated blocks which are representative for the Lochaline Sandstone. The core plugs were taken with the intent of minimal spacing between the 18 samples in form of one inch diameter core plugs to avoid complete utilisation of the sampled rock.

The samples were divided into 3 different series. Samples of series labelled with H were taken in perpendicular orientation to samples of series G. The core plugs of the O-series were obtained from another block. The number of the individual samples of one series does not represent the order of sampling.

Each sample was cut into a central part of 2 inch length and two off-cuts. After 2-3 days of drying at 60°C, the following techniques were employed on these samples (see also graphic in figure 6):

Utilisation of the first cut-off:

- Thin sections were prepared from the following samples (O6, G16, H1, H3) and examined by transmitted light microscopy using a Leitz optical microscope in order to obtain information about texture and modal composition. Additional thin sections from samples (O7, O8, G13, G15, G17, G18, G19, H2, H5) were prepared to measure point counting porosity.
- Thirteen polished thin sections were prepared for two dimensional porosity-analysis and pore size distribution by back scatter electron microscopy (BSEM), using an ISI ABT55 scanning electron microscope (SEM) coupled with an image analyser. The SEM equipped with an energy-dispersing X-ray analyser, in this case the Link Analytical EDS AN 10 55 system, also permits a qualitative elemental analysis of the minerals (EDX-analysis). After polishing off of the carbon coating, the cathodoluminescence of the samples were examined. The instrument used is a Citl Cold Cathode Luminescence 8200 mk3 with a gun current of 620-630 mA, in combination with an ordinary light microscope.
- Four samples were prepared for examination of their fresh broken surfaces, using the SEM in order to obtain surface textures, structures of three dimensional pore throats and information about their filling.
- Four samples were prepared for examination of their three-dimensional pore cast, using the SEM to gain information on the pore geometry and pore size distribution.
Utilisation of the second cut-off:
21 samples were analysed by a mercury injection test using Carlo Erba Instruments Macropore 120 Unit and Carlo Erba Instruments 2000WS Porosimeter. The raw data gathered from the tests was processed using the Carlo Erba Instruments Milestone 100 software and the MicroSoft software Excel. Finally the following data was obtained: pore radius distribution, porosity, specific surface, fractal dimension and grain size distribution. The limit for the maximal input pressure was adjusted to 199 MPa or 28861.5 psi.

Utilisation of the central core plugs (2” length, 1” diameter):
The porosity of 22 samples was measured by using a Coberly-Stevens Boyl’s Law porosimeter which is operating with He-gas. The analysis were performed at an input pressure between 95 and 97 psi, during each individual measurements the input pressure was kept constant.

On the same 22 samples the Klinkenberg Permeability was measured by using a permeameter which operates with N2-gas. The flow rates are ranging within 0.25 – 3.6 ml / sec. and the pressure gradient varies within 0.02 – 1.7 atm. The sleeve which contains the samples was adjusted to a constant pressure between 400 and 410 psi.

Electrical conductivity was measured on 16 samples at 1 kHz using a QuadTech 7400 Precision RLC Mete. The samples were completely saturated with a 1 molar NaCl solution and the sleeve which contained the samples was adjusted to a pressure of 400 psi. Dielectric properties were measured in the frequency range of 10 Hz – 500 kHz.
Fig. 6: Core sampling and the routine and special core analysis techniques employed

- **Light Microscopy**
  Porosity, Grain Size Distribution
- **Scatter Electron Microscopy (SEM)**
  Porosity, Pore Size Distribution
- **Gas/Brine Permeability**
  (C.Press: 400psi, Kt.-corrected)
- **Helium Porosity**
- **Calliper Porosity**
- **Mercury Porosity Curve, Distribution**

(C.Press.: 400psi, 100% sat., 1kHz)
III. Methodology

3.1 Sedimentary Logs at Beinn na h-Uamha
Sedimentary logs were drawn at five outcrops of the Lochaline Sandstone along a 1200m west-east transection at Beinn na h – Uamha. The sequence of the LSst was subdivided into four units in order to enable a correlation between the outcrops of different altitude. Special emphasise was drawn to the cemented horizons.

3.2 Core Plug Study
3.2.1 Mercury injection
Mercury injection was performed on all samples with the exception of sample G20. Mercury injection is a relatively quick method for gaining capillary pressure data from which a number of parameters can be calculated. In this study calculations were made of pore volume, pore radii at different injection pressures. As well bulk density, average pore radius, porosity, fractal dimension, specific surface (appendix 8.1) and grain size distribution of each sample was calculated.

A histogram has been produced to illustrate the porosity results (fig.33). Data plots of pressure, pore radii and mercury saturation per capillary pressure on the one hand versus mercury saturation on the other hand were produced to obtain an graphical impression of interdependent parameters (appendix 8.2). A histogram of the pore size fraction on a logarithmic scale versus mercury saturation enables also to visualise the amount of small pores (areas of blocks are not comparable because of the log presentation).

The upper limit for the determination of the pore size with the macropore unit is a radius of 30 µm, the smallest resolvable radius of 0.003 µm is given by the max pressure of 199 MPa (Kuhlenkampff, 1994).

Theory
During the experiment a non-wetting liquid (Hg) is entering the pores in the sample against their capillary pressure. The data which were obtained enclose the necessary external pressure which has to be applied to overcome the corresponding capillary pressure.

The external pressure required (p) is inverse proportional to the pore radius (r), its relationship is known as the Washburn equation:

\[ p = -c \left( \frac{2\sigma \cos \theta}{r} \right) \quad (1) \]

The surface tension of mercury (\( \sigma \)) of 480 dynes/cm has been taken as the average at 25°C. The wetting angle of the mercury (\( \theta \)) on the solid wall of the pore is 141.3°. C is a conversion factor (0.145 psi / (dynes / cm - micron))r is in µm units.
The Washburn equation is derived from the assumption that all pores are cylindrical and thus from the consideration of the following forces with contrary efficacy:

The force which is tending to push the liquid out of the capillary is perpendicular to the plane of the contact surface \(2\pi r \sigma \cos \theta\). Because of the fact that mercury has the tendency to spread on the pore wall surface, the cosine of the wetting contact angle completes the formula (by convention, the contact angles are measured through the wetting phase fluid):

\[
-2 \pi r \sigma \cos \theta = \pi r^2 p
\]  

(2)

The contact angle reflects the degree to which a surface is wetted by a fluid (its wettability).

Fig. 7: Graphically illustration of the wetting contact angle

Against this force, the external pressure will be exerted over the area within the contact circumference:

\[
\pi r^2 p
\]  

(3)

These two forces have the same value when the equilibrium is reached:

\[
-2 \pi r \sigma \cos \theta = \pi r^2 p
\]  

(4)

The fact that the surface tension \(\sigma\) and the angle of contact \(\theta\) are well known values allows to calculate the pore radius \(r\), in microns, if the capillary pressure \(p\), in psi units, is known:

\[
r = \frac{108.6}{p}
\]  

(5)

Procedure

The experiments were carried out on Carlo Erba Instruments Porosimeter. They are the Macropore Unit Model 120, the Porosimeter 2000WS and the Milestone 100 Software System.

For the prevention of fractures and coarse surfaces most of the samples were cut by a saw in an almost semi-cylindrical form of 0.2 - 0.8 cm length. The difference in sample size is unlikely to affect the predicted permeability accordingly to results of sandstone data presented by Kamath (1992). Then the samples were dried at 60-65°C for two days.

Neither for conformance artefacts nor for surface porosity artefacts were made corrections. The conformance artefact is generally insignificant and is caused by the energy (pressure)
required to conform the mercury meniscus to the sample. The more angular and irregular the sample the larger is this artefact, as proven by Bowen et al. (1995) on steel billets. An artificial surface porosity is produced during preparation of the sample, by cut through the pore network which opens pore bodies and creates additional pores through grain losses at the surface of the sample. The higher the "roughness" of the surface and larger the surface to volume ratio the higher this artefact. Since the samples of the highest surface roughness (O-series) represent also the samples with the largest porosity, the additional surface porosity is unlikely to affect the porosity measurement, but could affect the entry pressure.

**Measurement using the Macropore Unit**

After drying the weighed sample was placed inside the glass dilatometer. Then the dilatometer was evacuated for a few minutes until digital display reads close to zero and the holder position was adjusted. The vacuum switched off for one minute to verify the system is leak-proof. Under dynamic vacuum conditions (vacuum pump on) the dilatometer was filled drop wise up to the line mark (100mm Hg). Then the dilatometer was returned to atmospheric pressure in several steps.(the earlier the first measurement the better the resolution of larger pores). The penetration volume at each selected pressure was recorded by zeroing the cursor each time a new pressure was measured.

![Fig. 8: Measurement steps using macropore unit](image)

At the end of the measurement cycle the dilatometer was removed and its weight recorded (sample + Hg + dilatometer).
Measurement using Porosimeter 2000 (Micropore Unit)
Then the measurement already described was proceeded with higher pressure. The dilatometer was placed inside the autoclave and the autoclave was filled with high pressure oil. The important point in this step is to prevent air bubbles inside the autoclave. The monitoring system is based on the variations on the mercury level in the calibrated stem of the dilatometer. The progress of the penetration of the mercury into the sample is electrically followed by means of a patented capacitance system; when the pressure increases, the level of mercury in the stems falls, consequently the capacitance pressure. Pump stops allowing the collection of points pressure/volume according to the algorithm set by Carlo Erba Instruments. This algorithm regulates the functioning of the pump and when the detection of a minimum mercury penetration is achieved it will stop the pump temporarily allowing the stabilisation of the volume data reading. This delay time for reaching the equilibrium was set to 50 seconds.

At the control unit the maximum pressure was set to a value of 199 MPa (1990bar). The pump speed was 2, the decreasing cycle 2bar/sec.

Weak points of the method
In almost any porous substance there are no cylindrical pores. For irregularly shaped pores the ratio between the pore cross-section (related to the pressure exerted) and the pore circumference (related to the surface tension) is not proportional to the radius and depending on the pore shape, so the values of the radius will be lower. A further source of error concerns the topology of the way to a particular pore. If the way goes through smaller pores throats, the volume of down-stream bigger pores will count to a smaller pore range in the final distribution of pores (Jonas, 1988 in Kulenkampff, 1994). The capillary pressure hysteresis curves gives a possibility to quantify that effect, because the pores which were filled through smaller pore throats have no chance to evacuate their pores while the pressure decreases. Larson and Morrow (1981) revealed that a decrease of the fraction of less accessible interior pores by reducing the size of sample and thereby increasing the surface to volume will result in capillary pressure curves which are more closely related to a actual pore size distribution. Finally, it is thought that the calculated pore size distribution in this study, from capillary curves by employing the bundle of tube model, will underestimate both small and large pore throats. Moreover, I would like to cite Dullien (1992): “nothing could be further from the truth”. For example, fractures and the spaces between spherical particles cannot be described by a pore size. Also the wettability throughout “one pore” may not even be constant.

The further weak point of the technique is the irregular record of the pressure/volume readings according to an internal algorithm which controls the pump. Calculations proved that it is not a specific, regular amount of mercury penetration which results in a pump stop and a
pressure/volume reading. As an improvement appears the method of Yuan and Swanson (1989). They used a method of rate controlled porosimetry in which the mercury injection rate is kept constant and the pressure is monitored. Furthermore the mercury injection is a destructive test; the samples can not be used again for other measurements and finally, mercury form is a very health hazard.

Why is mercury injection still a popular method?
The advantages of the technique are that it is rapid (3-4h) and automatically and that irregular, small samples can be used (drill cuttings can provide data).

Additional parameters gained:
- Grain size determination
  The determination of the grain size includes the following equation during processing:

  \[
  D = \frac{\alpha \times [-203.75 \times \sigma \times \cos (\theta \times 0.01745)]}{1000 \times p} \quad (6a)
  \]

  \[
  D = \frac{454}{p}
  \]

  where
  \(
  \alpha = \text{compressibility factor } 4.65 \quad \sigma = \text{surface tension } 480 \text{ dynes/cm} \\
  p = \text{pressure in bar} \quad \theta = \text{contact angle } 141.3^\circ \\
  D = \text{grain diameter [µm]}
  \)

  It reveals an simple relationship between capillary pressure and grain size. The earlier the first pressure/volume reading were carried out, the better would be the resolution in the “coarser” grain size range.

- Porosity
  The effective porosity \(Hg\) was measured by the determination of the bulk sample volume and the pore volume (= volume mercury at maximal pressure):

  \[
  \frac{\varepsilon_{Hg}}{} = \frac{V_{pore}}{V_{Bulk}} \times 100 \quad (6b)
  \]

  where
  \(\varepsilon_{Hg}\) = Mercury porosity \(V_{Bulk}\) = Mercury bulk volume \(V_{pore}\) = Mercury pore volume


### Theory and Operations

Basically the whole measurement is based on Boyle’s Law, which is used to provide an estimate of grain volume. The gas law states that the product of pressure $P$ and volume $V$ of a given quantity of gas is proportional to the amount of gas occupying that volume:

$$ P \times V = P_1 \times V_1 \text{ at constant temperature} \quad (7) $$

where $P_1, V_1$ = corresponding values of pressure and volume under other conditions.

In the porosimeter the cores are placed in a chamber. This chamber is then filled with gas. From the change in pressure conditions of the gas after the emplacement of the sample, one can measure the matrix volume.

---

**Fig. 9: Plan of the Helium porosimeter**

The instrument consists of a reference cell with the volume $V_R$ and a sample cell with the volume $V_S$. The grain volume of the sample is termed $V_{\text{Grain}}$.

The reference cell is filled through the left valve with He-gas at a load pressure $P_0$ between 95 and 97 psi. After the reference cell is shut and the pressure is recorded, the sample cell is shut by closing the valve to the atmosphere. There are now two pressure-volume products:

$$ (P_0 + P_{\text{Baro}}) \times (V_R) = P_R \quad (8) $$
$$ (P_{\text{Baro}}) \times (V_S - V_{\text{Grain}}) = P_S \quad (9) $$

where

$P_S$ = Pressure in the sample cell  \quad $P_R$ = Pressure in the reference cell
From now on it was made the assumption that the barometric pressure $P_{\text{Baro}}$ was constant during one measurement cycle. Thus the “$P_{\text{Baro}}$” term could be ignored.

For the determination whether the assumption is accurate or not, the “zero” pressure was recorded at the “start” and “end” of each cycle. Then the centre valve is opened and the pressure stabilises after a few seconds at a lower pressure $P$. The sum of the pressure-volume products from both cells must equal the pressure-volume product of the gas in the whole system:

$$ (P_0 \, V_R) + (V_S - V_{\text{Grain}}) = (P) \, (V_R + V_S - V_{\text{Grain}}) \quad (10) $$

The last step is to solve the equation for the grain volume $V_{\text{Grain}}$ of the sample:

$$ V_{\text{Grain}} = V_R + V_S - V_R \left( \frac{P_0}{P} \right) \quad (11) $$

To make the measurement more accurate the term $V_S$ (volume of the sample cell) had to be eliminated. That is why the same operation was repeated by using a sample cell full of billets $B$, for which the volumes are known:

$$ B_1 + B_2 + B_3 = V_R + V_S - V_R \left( \frac{P_1}{P_2} \right) \quad (12) $$

The grain volume is independent from the sample cell volume $V_S$ by subtraction of eq.11 from eq. 12:

$$ B_1 + B_2 + B_3 - V_{\text{Grain}} = - V_R \left( \frac{P_1}{P_2} \right) + V_R \left( \frac{P_0}{P} \right) \quad (13) $$

(Now solution for grain volume $V_{\text{Grain}}$ possible)

By a third measurement, using a sample cell volume created by removing billets of a volume which is close to the expected pore volume of the sample in question, from the cell originally filled with billets the reference cell volume $V_R$ can be found:

$$ B_2 + B_3 = V_R + V_S - V_R \left( \frac{P_3}{P_4} \right) \quad (14) $$

Volume of the reference cell by subtract eq. 14 from eq.12:

$$ B_1 = V_R \left( \frac{P_3}{P_4} - \frac{P_1}{P_2} \right) \quad (15) $$

(Now solution for $V_R$ possible)

The pore volume is found by subtracting the grain volume obtained with the instrument from bulk volume. Because some samples are not very uniform right cylinders with flat ends (gaps
in the sample chamber would be counted as pore space) the bulk volume was measured by the
Archimedes method (see below) in which the apparent loss in weight of the saturated sample
is measured upon suspending in 1 M NaCl of known density. The porosity fraction was then
calculated by dividing the pore volume by the bulk volume:

\[ V_{\text{Bulk}} - V_{\text{Grain}} = V_{\text{Pore}} \quad (16) \]

\[ \He = \left( \frac{V_{\text{Pore}}}{V_{\text{Bulk}}} \right) \times 100 \quad (17) \]

where

\[ \He = \text{Helium porosity} \quad V_{\text{Bulk}} = \text{Archimedes bulk volume} \]

\[ V_{\text{Grain}} = \text{Helium grain volume} \quad V_{\text{Pore}} = \text{Pore volume} \]

Advantages-disadvantages of the method
It is a non-destructive, fast and accurate method to measure porosity but the measurement
requires core plugs.

Remarks:
Since there was not expected a significantly unconnected pore volume, the sample was not
crashed previously to the grain volume measurement.
High values of porosity could be due of warming up the sample cup, correspondingly high
grain values!
Low permeability samples require long equilibration times to allow diffusion of helium into
the pores. Failure to allow adequate time will result in high grain volume and so lower
porosity.

3.2.3 Archimedes Porosimetry
The method of Archimedes was applied because of the uncylindrical form of the core plugs
and hence the inaccuracy by measuring the bulk volume with a measuring stick. It was not
possible to improve the form by polishing the flat ends without obtaining impurities.
The principle of Archimedes is based on the following consideration: a body immersed in a
fluid experiences a resultant upward buoyant force equal in magnitude to weight of a
displaced fluid.

\[ F_B = \delta \times g \times V_{\text{Bulk}} \quad (18) \]

where

\[ \delta g = \text{Density of the fluid (1M NaCl)} \quad g = \text{earth’s pull (9.81 m/sec)} \]

\[ F_B = \text{Buoyant force} \quad V_{\text{Bulk}} = \text{Sample bulk volume} \]
<table>
<thead>
<tr>
<th>Bulk Volume / Area</th>
<th>Helium: $V_{\text{Grain}}$</th>
<th>Fluid Imbition: $V_{\text{pore}} + V_{\text{bulk}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>24500 mm$^3$</td>
<td></td>
<td>(Arch., Calliper)</td>
</tr>
<tr>
<td>~1000 mm$^3$</td>
<td>Mercury: $V_{\text{pore}} + V_{\text{bulk}}$</td>
<td></td>
</tr>
<tr>
<td>~54 mm$^3$</td>
<td>Point Counting: Pore Area</td>
<td></td>
</tr>
<tr>
<td>~13.5 mm$^3$</td>
<td>Image Analysis: Pore Area</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 10: Porosity measurement: different scales and techniques
The net buoyant force $F_B$ in equilibrium result in following equation according to Newton’s first law (the sum of the forces $F$ must be zero):

$$ F = F_B + T - m_{sat}g = 0 \quad (19a) $$

respectively

$$ \delta \cdot g \cdot V_{Bulk} = m_{sat}g - T \quad (19b) $$

where

$T = \text{Tension in the string (weight immersed)} \times 9.8 \text{m/sec.}$  
$g = \text{Earth’s pull (9.81 m/sec)}$  
$m_{sat}g = \text{Weight saturated with fluid} \times 9.8 \text{ m/sec.}$  
$\delta = \text{Density of the fluid (1M NaCl)}$

The volume can be calculated from saturated weight of the sample substracted by the immersed weight of the sample and divided by the known fluid density:

$$ V_{Bulk} = \frac{(m_{sat}g - T)}{\delta \cdot g} \quad (20) $$

The grain volume or the dry weight is needed to determine porosity. The weight of the dry rock substracted by the weight of the saturated rock results in the weight of the fluid in the pore. Since the fluid density is known and the weight of the fluid in the pores, it is a simple calculation to obtain the pore volume which contains this fluid and then the porosity:

$$ V_{Pore} = \frac{(m_{sat} - m_{dry})}{\delta} \quad (21) $$

$$ Arch = \frac{V_{pore}}{V_{bulk}} \times 100 \quad (22) $$

where

$Arch = \text{Archimedes porosity}$  
$m_{dry} = \text{Weight of the dry sample}$  
$m_{sat} = \text{Weight of the saturated sample}$  
$\delta = \text{Fluid density}$
3.2.4 Calliper Porosimetry
The calliper method is based on the bulk volume measurement of the core plug with a measuring stick. The pore volume is determined by subtraction of the saturated weight from the dry weight; saturation with 1 M NaCl is achieved under vacuum. Because the density of the fluid is known the volume of the fluid in the pores can be calculated (pore volume).

\[ \text{Cal} = \frac{V_{\text{Por}}}{V_{\text{Bulk}}} \times 100 \]  

where
- \( \text{Cal} \) = Calliper porosity
- \( V_{\text{Por}} \) = Calliper pore volume
- \( V_{\text{Bulk}} \) = Calliper bulk volume

3.2.5 Gas Permeability (Klinkenberg corrected)
The measurements were carried out by placing the samples in Hassler sleeve core holder at low confining pressures of about 400 psi to prevent gas flow around the plug. Then a nitrogen pressure was applied on one end. The flow rate was measured by means of a calibrated outlet (atmospheric). The upstream and downstream pressures were indicated by gauge pressures to which was added the atmospheric pressure to determine the real absolute pressure.

Darcy’s law forms the base for the measurements:

\[ K = \frac{\mu \times Q \times L}{A \times (P_1 - P_2)} \]  

The definition is wholly empirical. The equation expressed in words means: If a liquid having a viscosity of 1 (\( \mu \)) flows through a porous rock of 1 cm length (L) and 1 cm² cross-section (A) at a rate of 1 cm³/sec (Q) when the pressure differential between input and output is 1 bar then the rocks permeability would be 1000 millidarcy. Thus permeability is associated with the flow of fluids through the rock.

Because of the fact that fluid could react with the rock, the measurements were carried out with \( \text{N}_2 \)-gas.

The flow rate of gas was measured due to the following relationship:

\[ P_1 \times Q_1 = P_2 \times Q_2 \]  

\( P_1 \) and \( P_2 \) are the input and output pressures, \( Q_1 \) and \( Q_2 \) are the corresponding flow rates.

Because \( P_2 \) was known as the atmospheric pressure, the Darcy relationship for gases looks as following:

\[ K = \frac{2\mu \times Q \times L \times P}{A \times (P_1 - P_2)^2} \]  

where
- \( P_1 \) = absolute input pressure
Methodology

P₂ = absolute output pressure
P = atmospheric pressure
Q = rate of air flow measured at atmospheric pressure

Klinkenberg correction

Using gas at low pressure leads to an apparent permeability which is too high because the mean free path of the gas is no longer negligible compared to the typical pore size. (Klinkenberg effect, fig. 12). Corrected permeability values are obtained by measuring the permeability at series of different mean pressures. Klinkenberg plots (appendix 8.5) of permeability versus reciprocal mean pressure are made by the extrapolation of the measured data on a straight trend line to infinite mean pressures. The slope of the trend line is called the Klinkenberg gas slippage factor. The extrapolated point determines the theoretically permeability equivalent to a permeability which would be obtained if flowing a non-reactive liquid (oil). This corrected permeability is the so called Klinkenberg permeability Kₜ.

No correction was employed for the Forchheimer effect at turbulent flow.

Note: Gradual compaction of poorly cemented samples can occur even with sleeve pressures as low as 400psi (BP Core Analysis). The precision depends upon permeability and is stated to be in the following order (Shell Core Analysis):

1 - 50 mD accuracy: 10 %
50 - 2000 mD accuracy: 5 %

Trend lines should not be overvalued for the assessment (R²) due to the difficulty to choose the right number and values of the input pressures.

The investigation of the pressure dependence of permeability were carried out without back pressure. Half an hour was chosen as the sufficient consolidation time between each pressure step and the permeability reading.

![Fig. 12: Klinkenberg effect](image-url)
3.2.6 **Probe Permeability**

Permeability measurements with the probe permeameter are carried out in situ in the Lochaline mine in section N-S 13.

In probe permeametry gas of known pressures is injected from the probe to the sample rock, the gas flows from the end of a small-diameter tube that is sealed against the rock surface. The pressure in the probe is measured together with the corresponding volumetric gas flow rate. The gas permeability is determined from calibrations based on pressure and flow rate.

To obtain values which are close related to the absolute permeability values, the maximum Nitrogen gas injection pressure should be keep constant during the outcrop measurements and the flow rates and permeability values should be calibrated in the lab under the same conditions as in the field. For the calibration is a measurement of flow rate versus injection pressure at a set of at least 10 homogeneous samples with known laboratory permeability values necessary. The plot and the resulting equation from the linear regression is shown in figure 13. The probe permeability (mD) can be calculated from the normalised flow rates measured in centimetres cubed per minute.

![Calibration Normalised Flowrate versus Permeability](image)

Fig. 13: Trendline and equation used for calibration of the probe permeameter

During the measurement itself it is important to seal the contact between rock and probe permeameter to obtain constant gas injection rates. The gas tube should be pressed perpendicular to the flat rock surface under adequate pressure. Leaking is easy to be recognise by whiz of gas. The pressure at which the flow rate was taken should be written down, to record any change during measurement process. In order to avoid any influence of weathered surfaces, all measurements were carried out on an grid of sampling points of already prepared “fresh” flat surfaces.

The advantage of the technique is the undisturbed insitu measurement without any influence of rock preparation and the relative easy and fast handling. Certainly, compared with laboratory determination of the permeability, it is only useful in determination of a relative trend in permeability. The data may reflect effective permeability at a partial fluid saturation.
3. 2. 7 Brine Permeability

The brine (1 M NaCl) permeability was determined in the comparable configuration to the gas permeability, at 500 psi confining pressures, for the samples H3, G17, G19, O2, O3 and O4. It was applied a pressure gradient and the flow rate was measured manual. The bulk of the applied flow rates were ranging between 3 and 9 cc/minute. The final permeability values of each sample are calculated as averages permeability values from three different flow rates employing the following equation:

\[ K = 14700 \frac{L}{A} \frac{v}{Q} \frac{1}{\Delta P} \]

where,
- \( K \) = permeability [mD]
- \( L \) = Length of the core plug [cm]
- \( A \) = Area of the core cross-section [cm²]
- \( v \) = viscosity [cp]
- \( Q \) = flow rate [cm³/sec.]
- \( \Delta P \) = pressure gradient [psi]

3. 2. 8 Electrical Resistivity

Digression into the definition of parameters

In the following is given short definition of conductivity, formation factor, m-value, tortuosity and an introduction of the calculation of dielectric properties.

Formation Factor and Conductivity

The main contribution to conductivity in rocks is generally made by electrolytes in their pore space rather than by the rock matrix. These fluids (in our case NaCl-solution) can contribute to conductivity in two ways: either by their intrinsic electrolytic conductivity or by their electrochemical interaction with the solid material at their interface. In the first case the pore space determines the geometry of the conductive path, in the latter the internal surface along pores and cracks. The complex conductivity of a saturated rock is therefore assumed “to be related to its porosity, pore geometry, and the nature of the surface of the mineral grains lining the pores, as well as the dielectric properties of the mineral grains and pore fluid” accordingly to Glover et al. (1994). The conductivity of a saturated porous rock can be described by the following equation:

\[ \sigma = \left( \sigma_w / FF' \right) + \sigma_s \]  \hspace{1cm} (27)

Here \( \sigma \) is the bulk rock conductivity, \( \sigma_w \) the electrolyte conductivity, \( \sigma_s \) the surface conductivity and FF' the “true” formation factor, a rock specific micro structural constant. The determination of the “true” formation factor requires a series of at least 6 measurements of bulk rock conductivity at different electrolyte concentrations (e.g.: 1 ppm 0.00001 ppm). The slope of the curve at higher conductivity’s in a double logarithmic plot will determine the “true” formation factor. If the second term of equation 27 is negligible compared with the first
term, the equation is reduced to (Sundberg, 1932):

$$\sigma = (\sigma_w / FF)$$  \hspace{1cm} (28)

Thus the bulk rock conductivity will be proportional with the conductivity of the electrolyte in the pore system, reduced by the factor $1 / FF$ (now FF is not longer a rock specific constant, it depends among others also on the electrolyte). For a better understanding of the meaning of the formation factor equation 27 will be divided by $\sigma_w$:

$$1 / FF = 1 / FF' + \sigma_s / \sigma_w$$ \hspace{1cm} (29)

It is now clear that the formation factor FF depends on the microstructure ($FF'$) and the ratio of electrolyte to surface conductivity. Since in most cases the electrolyte conductivity is several orders bigger than the surface conductivity, the second term could be again ignored:

$$FF \approx FF'$$ \hspace{1cm} (30)

The following two extreme cases outline the tightly inverse connection between formation factor and porosity, which is on a closer examination in the next paragraph. If the porosity is infinite large, the bulk conductivity and the electrolyte conductivity are equivalent and the formation factor in equation 28 would have an approximate value of one. If the porosity decreases, the ratio of electrolyte to bulk rock conductivity in equation 28 will increase, thus the value of the apparent formation factor will increase (as well as the true formation factor, equation 30). If the true formation factor is increasing, the value of the ratio electrolyte / true formation factor in equation 27 becomes smaller and the surface conductivity as part of the bulk conductivity becomes more important.

**Tortuosity and Cementation Factor (= m-value)**

The tortuosity of a pore system in rocks is theoretically defined as the ratio of the mean path length $A$ to the straight line distance $B$ of the overall path in the sample (fig.14). Clearly, the tortuosity value is always $>1$ in natural porous media. It includes all factors that alter the transport in the porous rock including pore-throat constrictions, isolated pores, dead-end pore paths and even grain surface effects. Measurement of tortuosity is normally performed indirectly by comparing the electrical conductivity of an electrolyte solution in a porous rock with the corresponding conductivity of the pure pore solution (equation 28). The electrical conductivity ratios yield a parameter already termed formation factor, which has also been
defined by the equation (Wyllie, 1957):

$$F = \frac{\tau^2}{\phi} \quad (31)$$

Here $\tau^2$ is the tortuosity.

Formation factors are commonly estimated using the empirical relationship (Archie, 1942):

$$F = \phi^{-m} \quad \text{Archie’s First Law} \quad (32)$$

Archie (1952) reported that $m = 2$ for packet sand and $m = 1.9$ for consolidated sandstones. But often is observed that the $m$-value increase with packing of grains, compaction and degree of cementation of rocks and usual values are in the range of 1.3 - 2.5 (Landolt Börnstein, 1982). Further factors could also affect the $m$-value: type of porosity, tortuosity, pressure, wettability, pore geometry’s and clay content, summarised in Timur et al. (1972). For crystalline rocks were observed $m$-values consistent with Archie’s Law (Brace et al, 1965). But more recently published data (Oelkers, 1996) reveal also a considerable scatter in crystalline rocks. Nevertheless formation factors generated from Archie’s Law can be still seen as a first approximation, but one should be aware of the uncertainties of at least +/- one order of magnitude.

A further possibility to obtain the formation factor is the ratio of specific surface to surface conductivity (and other factors themselves connected with the internal surface: permeability, electrochemical potential, cation exchange capacity etc.). Beside empirical confirmation, there exists also theoretical justification for such relations (Revil et al., 1997).

![Graphically description of the tortuosity](image)

**Fig. 14:** Graphically description of the tortuosity as the ratio of the length of conducting path A to the length of straight line B.

**Calculations of Dielectric Parameters**

For an assumed electrical sinusoidal current, the resistance measured could be splitted into the imaginary and the real part of the complex resistance, respectively $X_s$ and $R_s$. The first step is to convert this measurement into the real ($\rho'$) and imaginary ($\rho''$) part of the specific complex resistivity ($\rho$) by scaling by the physical dimensions (length $L$ and area $A$) of the cylindrical samples.

$$\rho'(\omega) = R_s(\omega) \times A / L \quad (54)$$
\[ \rho''(\omega) = Xs(\omega) \ast A / L \quad (55) \]
\[ \rho(\omega) = \rho'(\omega) + \rho'' \quad (56) \quad (\omega = \text{function of angular frequency}) \]

The current flow could certainly also be described by conversion into the complex conductivity \( \sigma \):
\[ \sigma'(\omega) = \rho'(\omega) \ast \left[ 1 / (\rho'^2(\omega) + \rho''^2(\omega)) \right] \quad (57) \]
\[ \sigma''(\omega) = \rho''(\omega) \ast \left[ 1 / (\rho'^2(\omega) + \rho''^2(\omega)) \right] \quad (58) \]
\[ \sigma(\omega) = \sigma'(\omega) + \sigma''(\omega) = 1 / \rho(\omega) \quad (59) \]

The second step is to reveal any frequency dependence of the complex conductivity, which defines the permittivity \( \kappa \), normalized by the dielectric permittivity of the vacuum \( \kappa_v \):
\[ \kappa''(\omega) = \sigma''(\omega) / \omega \ast \kappa_v \quad (60) \]
\[ \kappa'(\omega) = \sigma''(\omega) / \omega \ast \kappa_v \quad (61) \]
\[ \kappa(\omega) = \kappa'(\omega) + \kappa''(\omega) \quad (62) \]

It should be noted that \( \kappa \) is also known as the dielectric constant, because at high frequencies energy dissipation is negligible and so \( \kappa \) becomes a constant number. It should also be noted that the real part of the complex relative permittivity \( \kappa' \) consists of the a converted imaginary part of the complex conductivity \( \sigma'' \).

Frequency dependence is a result of rock behaving both as a conductor and as a dielectric (insulator). There are much fewer ionic charge carriers in the matrix material than in the electrolyte and thus displacement processes in response to an electrical potential gradient are diffusive. Certainly the electrical current under an applied electric field will almost totally flow through the higher conductive pore fluid, in our case NaCl-solution. An applied field at relative low frequencies, will charges allow time to migrate through the pore system. If such migrating ion’s accumulate at solid interfaces (due to the movement of anions and kations in opposite direction), it leads to large interfacial polarisations and \( \kappa^-\)values. Such a charge redistribution effect, also called Maxwell-Wagner effect, could produce even larger dielectric constants than either that of quartz or fluid alone. Whereas polarisation effects in the very low frequency range (<1kHz) between electrodes and the electrolyte in the rock are not desired, space-charge polarisation, the predominant effect in the examined frequency range from approximately 10kHz - 500kHz, reveal useful information about the rock microstructure.

Cole-Cole (1941) introduced a modified Debye-model to account for a frequency dependence of the complex conductivity in a porous media:
\[ \kappa(\omega) = \kappa(\infty) + \left[ \kappa(0) - \kappa(\infty) / 1 + (\omega \tau)^{1-q} \right] \quad (62) \]

\((1 \geq q \geq 0)\)

where \( \kappa(\infty) \) and \( \kappa(0) \) are, respectively, the values at zero and infinite frequency. The dielectric constant \( \kappa \) decreases from its lowest frequency value \( \kappa(0) \) to its highest frequency value \( \kappa(\infty) \), where the latter is mainly a function of density (Gueguen and Palciauskas, 1994) and so of the
sample’s porosity. The parameters $\tau$ and $q$ describe a broad distribution of relaxation times associated with an ensemble of relaxation processes. Double logarithmic plots of $\kappa'$ and $\kappa''$ versus the frequency were used to show the dependence of the complex relative permittivity $\kappa$ over the whole frequency range measured. The $q$-exponent could be easily determinable from this plot by taking the gradient of the real part of the dielectric permittivity $\kappa'$ at high frequencies ($>10$ kHz), since the Cole-Cole behaviour of the real part of the permittivity varies with $-q$ exponential to $\omega$ (Ruffet, 1991). But in the low frequency range ($<10$kHz) exists a strong enhancement of $\kappa'$ and it is thought that the increase of the dielectric dispersion could be related to the shape of grains and so indirect to the Archie exponent $m$ (Kenyon, 1984).

The Cole -Cole response function could also be used to describe the complex resistivity:

$$\rho(\omega) = \rho(0) / [1 + (\omega\rho(0)\epsilon_c)^{1-q}] \quad (63)$$

(apparent permittivity $\epsilon_c \approx \text{square root of the conductivity}$)

The equation describes closest the plot of empirical $\rho'$- versus $\rho''$-values in the high frequency range, presented as a depressed semicircle in an Argand. It is also possible to obtain from an Argand diagram the $q$-value using the relationship $q = \alpha / \pi$, where $\alpha$ represents the angle between $\rho'$-axis and the centre point of the dispersion circle, but this method is much more inaccurate (especially if there exist only a small part of the circle). The physically “meaningful” values of the phase angle $\alpha$ are in the range of 0 - 0.5, based on physical theories and empirical data (Kuhlenkampff, 1994).

But the empirical Cole-Cole description is not satisfactory from a theoretical point of view. It is not only suggested that the Cole-Cole exponent could be replaced by a more complex functional factor but also that it is a function of a whole series of other physical parameters as well (Glover et al., 1994). Despite the theoretical point of view is not fully developed yet, empirical data proved, that a surface dependence of the Cole-Cole exponent exists at specific surfaces $\geq 1$ $\mu$m$^{-1}$ and that no clear relation between Cole-Cole exponent and degree of salinity of the rock fluid could be assumed (Kuhlenkampff, 1994), the latter in contrast to Glovers results (1994).

At low salinities the Cole-Cole exponent is very precise determinable, whereas at higher salinities and increasing pore volume conductivity the Cole-Cole exponent is less precise determinable. This is caused by the fact that the dispersive behaviour described by the Cole-Cole model contributes more to the bulk conductivity at low salinities, where surface conduction is the predominant process. Thus at a relative high electrolyte concentration of 1m NaCl, it is expected to be observed a dispersed behaviour only at a small frequency range.
**Equipment, Measurements and Calculations**

**Sample Preparation and Equipment:**
After drying, the samples were several hours saturated under vacuum with 1 ppm sodium chloride brine. Samples were also saturated for a second run within a fluid flow rig. This made it necessary to set a limit at which it is ensured that 100% of the rock is saturated. A time at a certain flow rate, equivalent to a volume flow which is 10 times bigger than the sample specific rock volume plus the volume of the wire between reservoir and core holder, was thought to be sufficient. Confirmed was that assumption by the later record of the rock resistivity during passing successions of different flow concentrations.

An electrical contact between rock and electrodes with minimised polarisation effects was ensured by using of fluid saturated filter paper (same fluid concentration) and platinum wire mesh at each end of the core plug. In order to avoid any “surface” conduction along the rough core surface, the cores were taped along their length axis.

Complex resistivity measurements were made using a QuadTech 7400 Precision RLC Meter with a basic measurement accuracy of 0.05%.

**Measurements:**
All conductivity measurements were carried out on rocks completely saturated in vacuum with NaCl brine at a confining pressure of 400 psi and at a frequency of 1 kHz. Conductivity measurements as a function of the rock properties were made at 1 kHz, since the phase angle is closest to zero and electrode-rock polarisation are not to be expected at that frequency.

The conductivity of a selection of samples (H2, H3, H5, G13, G14, G20, G21 and O1-O8) was measured after repeated saturated in the new installed fluid flow rig. Samples of all three series were measured at a frequency ranging from 10 Hz to 500 kHz to estimate any polarisation effects and to calculate the apparent fractal dimension from the real and imaginary part of the conductivity in the upper frequency range.

The conductivity of samples H5 and O8 was measured as a function of various electrolyte concentrations (from 2 mol to distilled water) to study grain surface conduction. The resistivity curve as a function of time during displacement of different concentrated pore fluids permits quantitative statements concerning the connected pore volume. It should be mentioned that a not to negligible amount of small white particles was transported by suspension out of the core during the saturation process.

The electrolyte conductivities were measured under the same conditions as the samples. Further should be noted that the electrolyte conductivity was only determined for the original electrolyte, but there may have been some change in electrolyte salinity due to rock electrolytes during the test handling.
Calculations:
Since the sandstone is assumed to be nearly clay-free and the electrolyte conductivity is relative high (1mol NaCl), no surface conductivities are expected. Therefore equation 28 was applied to determine the formation factor. Log-Log plots of bulk rock against electrolyte conductivity of the sample H5 and O8 were used to examine if there exists any surface conduction marked by a snap off of the curve at lower conductivities. Qualitative could any surface conduction be obtained as a first approximation by using of equation 27.

The m-value and tortuosity for each sample was determined by applying equation 32, or respectively 31, referring to the helium porosity. From the gradient of a log-log plot of formation factor versus helium porosity, the average m-value for the entire Lochaline Sandstone Formation was determined. Cross plots of m-values, tortuosities, permeabilities, porosities and formation factors were drawn to reveal any trends.

The complex resistivity and permittivity are calculated with equations 53 and 54 respectively 59 and 60.

3.2.9 Methods to Determine Fractal Dimension

Fractal dimension – what is it?
The rough surface structure of sandstone grains is too complex to describe it with conventional Euclidean forms. But most of them could by described by using of a fractal model which is characterised by the fractal dimension. In general the fractal dimension is defined by the equation (Mandelbrot, 1977)

\[ D = - \left[ \frac{\log N}{\log v} \right] \]  

(33)

where N is the number of substructures sitting upon a previous structure and 1/v is the size ratio between a substructure and the next larger one, thus the resulting structure is self similar. Any fractal structure can have any value between 0 and the Euclidean dimension in which the object is embedded. In our case is the grain surface embedded in the three dimensional pore volume. Crystal growth and dissolution processes are assumed to be the main contributor for the characteristic roughness of the grain surface, the higher the diagenetic alteration the higher should be the fractal dimension. Once the fractal dimension is determined it was used in the following way:

- Since transport properties (e.g. permeability) are unlikely to be related to the part of the rough surface, the fractal part of the porosity could be excluded from calculations of transport properties
- Attempting to refer the samples different degree of alteration to its fractal dimension
- Comparison of fractal dimensions measured with different techniques
- Assessment of the data measured: The lowest resolution limit associated with the fractal dimension can be related to the lowest resolution limit of the techniques used
Fractal structures are self similar, as defined above, that means they are invariant at different magnifications. A fractal structure comes across vividly by synthesis of a fractal model, e.g. the Van Koch curve (fig. 15). To construct such curve a geometrical structure of a initiator have to be defined (e.g. straight line-A) and also the geometrical structure of a generator (e.g. broken line-B). At every following, e.g. (k+1)st generation, each straight line section of the kth generator will be replaced by a scaled down version of the generator.

The length of the curves of each generation can be calculated in the following way: If the initiator line length is \( L_0 \) (= 0th generation, length 1), \( n \) the number of the smaller segments of the generator, \( v \) the factor of reducing of the previous generation and the generation in question is the first-one (\( L_1 \)), the equation to obtain the length would be:

\[
L_1 = L_0 \times \frac{n}{v} \quad (34)
\]

or

\[
L_{(1/3)} = 1 \times \frac{4}{3} \quad (35)
\]

That means that the first generation will consist of 4 smaller segments of length 1/3, that would be 4/3. That length is measured with the resolution of the length 1/3, the length measurement is therefore scale controlled.

The \( k \)th generation would then consist of the \( 4^k \) segments of length \( 1/3^k \):

\[
L_k = L_0 \times \left[ \frac{n}{v} \right]^k \quad (36)
\]

or

\[
L_{(1/3^k)} = \left( \frac{4}{3} \right)^k \quad (37)
\]
In case of the example of the Koch curve in figure 15, we could replace \( L_0 / v \) at the \( k \)th generation by \( \varepsilon \).

\[
\varepsilon = \frac{1}{3^k} \quad (38)
\]

(in other words: we adjust the scale of resolution \( \varepsilon \) to the length \( 1/3^k \))

That way \( k \) could be replaced by \( -(\log \varepsilon / \log 3) \).

\[
L(\varepsilon) = \varepsilon \cdot 4^{-\left(\frac{\log \varepsilon}{\log 3}\right)} \quad (39)
\]

because

\[
4^{-\left(\frac{\log \varepsilon}{\log 3}\right)} = \varepsilon^{-\left(\frac{\log 4}{\log 3}\right)} \quad (40)
\]

one could also write:

\[
L(\varepsilon) = \varepsilon \cdot \varepsilon^{-1\left(\frac{\log 4}{\log 3}\right)} = \varepsilon^{1-\left(\frac{\log 4}{\log 3}\right)} \quad (41)
\]

\[
L(\varepsilon) = \varepsilon^{1-\left(\frac{\log 4}{\log 3}\right)} \quad (42)
\]

It is now clear that the length of the line measured depends only on the scale of the resolution. The relation between scale and length is determined by the fractal dimension, in the case of the Koch curve:

\[
D = \log 4 / \log 3 = 1.26 \quad (43)
\]

The Koch curves can also be applied to a three dimensional model consisting of closed uniform Koch curves. The surface dimension will be 2.26 while the pore volume will have an Euclidean dimension of three.

There exists basically two different lines of research concerning the value of the fractal dimension of a rock pore system:

the Clausthal school with fractal dimension of maximal 2.36 derived from the pigeon-hole model (Pape, 1987a and 1987b) and higher fractal dimensions of 2.5-2.85 from analysis of fracture surfaces (Thompson et al., 1987), or dimensions of 2.55-2.95 from small-angle neutron scattering (Wong et al., 1984).

The first theoretical approach of modelling fractal surfaces in rocks, the pigeon-hole model, was presented by Pape et al. (1982). It is a modified Van Koch construction, based on semicircles. Assuming the growth of minerals in a pore system is restricted to the interface of the minerals, they proved that a maximum internal surface will be reached at input parameters of \( N = 26.5 \) and \( v = 0.25 \). Because they consider no increase in internal volume, a gap exists between the fractal dimension of 2.36 and 3. Any values between 2 and 2.37 should indicate a smoothing of the surface (Pape et al., 1987b).

In the following is a short description of models used to obtain the fractal dimension presented in this study:
Fractal models and techniques used to determine D

A. Mercury Injection / Menger sponge model

The fractal dimension of the surface can be determined by the measurement of pore volume $V$ as a function of pore radius $r$, applying the Menger sponge model (Friesen and Mikula, 1987). The model starts from a unit cube (initiator) subdivided into identical $m^3$ smaller cubes. A number of this cubes are removed accordingly to a given prescription, leaving a certain structure of cubes (generator). If we take the generator as a modified face of the cube (rather than a reconstructed), the object becomes fractal:

Again, we starting from a cube and are subdividing each face to $m^2$ smaller squares and removing $n$ sub-squares subject to the following restrictions: neither they should have common sides nor they should lie on the face’s edge (mathematically expressed as: $n^2(m^2 - 4m + 5)$. At each missing square one should insert a cube with a top face missing (net increase of surface: $4n$). This lead to an increase of the surface of the original face from 1 to $N_1/m^2$, where:

$$N_1 = m^2 + 4n \quad (45)$$

($N_1$ = generator)

After applying the generator at each face of the cube and repeating the procedure iteratively the fractal dimension of the surface becomes:

$$D = 2 + \log \left[ 1 + \left( \frac{4n}{m^2} \right) \right] / \log m \quad (46)$$

![Menger sponge model](image)

Fig. 16: Menger sponge model after two iterations, with $m=3$ (from Friesen and Mikula, 1987).

In order to determine the fractal dimension of a pore-grain interface from the change in pore volume during mercury injection, one could now express the pore volume of the above model in terms of the fractal dimension:

In the first step the pore volume on one faces becomes:

$$V_1 = \frac{n}{m^3} \quad (47)$$
Methodology

after two steps:
\[ V_2 = V_1 + \frac{nN_1}{(m^3)^2} \]  \hspace{1cm} (48)

after k steps and the summarising of the geometric series:
\[ V_k = V_{tot} \left[ (1 - \frac{N_1}{m^3})^k \right] \]  \hspace{1cm} (49)
\[ V_k = V_{tot} \left[ (1 - \frac{r_k^{-3-D}}{m^3}) \right] \]  \hspace{1cm} (50)

where the total number of cubes of size \( r_k \) is equal to \( 1/m^k \) and the total pore volume for the infinitely iterated structure \( (k \to \infty) \) is
\[ V_{tot} = \frac{n}{m^3 - N_1} \]  \hspace{1cm} (51)

By differentiating, the continuum limit of equation 50 yields to:
\[ \log \left( -\frac{dV}{dr} \right) \approx (2-D) \log r \]  \hspace{1cm} (52)

Instead of the radius calculated from the Washburn equation it was used the related raw pressure data, the final equation is therefore (after combining the above equation with the Washburn equation 1):
\[ \log \left( \frac{dV}{dP} \right) \approx (D-4) \log P \]  \hspace{1cm} (53)

The total pore volume at a given pressure is then given by the volume of mercury forced into the pores. The surface dimension of the sandstone samples are easily obtained from the slope of the log-log plot of \( dV / dP \) versus \( P \). The plots are divided into three groups accordingly to the pressure <1.2 bar, \~10 bar - 200 bar and > 200 bar. The gradient of the intermediate pressure plots is thought to represent surface fractal dimensions accordingly to previous measurements performed on coals (Friesen and Mikula, 1987), whereas values over 1000 bar were not used since they appear to be beyond the resolution limit of the aperture (large scatter of data points).

It is thought that at low pressures the mercury intruded rapidly into the interparticle pore system, whereas at intermediate and high pressures the mercury is forced into the pores of the pore-grain interface.

There could be a certain inaccuracy in the determination \( V(r) \). If there are any large pores which have only access through smaller throats, the intruded volume would count to smaller pore radii. It should also be noted that the range of possible surface dimension of 2 - 2.37 of the model (for max D: \( m=7 \) and \( n=13 \) in eq.46) is close to the maximal surface dimension of the Pigeon-Hole model. It should be noted that it is not possible with this method 0to distinguish whether the fractal dimension is derived from a fractal surface or a fractal volume.
B. Resistivity measurement (Model of Wang, of Ruffet and of Le Mehaute/Crepy)

Since the frequency dependence of the complex conductivity is caused by interface effects, it is possible to characterise the internal surface area and thus any related fractal surface dimension from electrical measurements. There already exist three models to obtain fractal dimensions (Le Mehaute and Crepy 1983, Wong 1987 and Ruffet et al. 1991) which are based on the empirical Cole-Cole model (Cole and Cole, 1941). Nevertheless, any fractal dimension obtained from those models should be seen only as an apparent fractal dimension, since the Cole-Cole exponent itself is also a function of electrolyte concentration and the scale of resolution (size of ionic type in solution) (Glover et al., 1994).

Description of Ruffet’s (R) model and the model of Le Mehaute and Crepy’s (MC):

The two models are described together, since both of them are based on the same physical assumptions. Their difference lies only in the different way of conversion the model into a two dimensional model. The first Model of Ruffet is given by,

\[ DR = \frac{2}{1 - q} \]  \hspace{1cm} (64)

the second equation describes the model of Mehaute and Crepy:

\[ D_{MC} = \frac{1}{1 - q} + 1 \]  \hspace{1cm} (65)

They assume that the dispersion is caused by the movement in two dimensions of ion’s along a fractal interface or rather caused by “the difficulty that conducting ions have in overcoming forces at the edges of plates of mineral surfaces that represent the pore surface” accordingly to Glover et al. (1994). Thus they assume that the interface of minerals is independent of the scale of analysis in the space of frequency in contrast to the scale dependence of other physical measurements of the interface.

Description of Wong’s model (W):

Wong’s model based on the assumption that ion's diffuse along an interface which is rather self affine then self similar, thus the pore surface is considered to behave as a self affine fractal. The relative permittivity is suggested to have the same frequency dependence as a polarisation that is assumed to be proportional to an average “drift” of an ion along the interface. The drift itself should be related to the diffuse distance with the exponent of D - q (D = Euclidean dimension).Thus the calculation of the fractal dimension is given by:

\[ DW = 3 - 2 \times q \]  \hspace{1cm} (66)

Taken the physically meaningful range of phase angles \( \alpha \), \( 0.5 \geq \alpha \geq 0 \), accordingly to Won’s
model the resulting dimensions would consist of values between 2.36 at high angles and, respectively, Euclidean dimension 3 at low angles. It should be noted that the values of the lowest dimensions at high angles of $\alpha$ are the upper limits of fractal dimensions obtained by the Pigeon-Hole and Menger-Sponge model.

In comparison using the theory of Le Mehaute and Crepy or of Ruffet the fractal dimension approaches 2 at low $\alpha$-values. At high phase angles the surface fractal dimension approaches 2.46, calculated by the Le Mehaute and Crepy Model, or respectively 2.93 using the theory of Ruffet.

Therefore the fractal dimensions are reverse related to the phase angle, in the two different physical models.

Since it is empirical proven that the phase angle is positive related to roughness surface area (Kuhlenkampff, 1994) and the specific surface which measures such surface roughness is assumed to be positive related with the fractal dimension (Pape, 1987b), it is expected that the fractal dimension increase with higher specific surface of a sample. Whereas fractal dimensions derived from the theory of MR and R are consistent with those assumptions, the reverse is observed for dimensions calculated with the model of Wong (Ruffet, 1991). Nevertheless, fractal dimensions calculated with the model of Wong show physical behaviour in relation to changes of salinity of the pore fluid. An increase in fractal dimension with decrease in NaCl concentration is expected and observed (Glover et al, 1994), since the surface conduction contributes more to the bulk conduction at lower fluid concentration and so the “resolution” scale to measure the surface roughness is much higher. Comparison of fractal dimensions obtained by applying the three models on experimental data sets shows that the values lies in a realistic range between 3 and 2 (Glover et al., 1994 and Ruffet et al.,1991)

C. SEM - image analysis

From images of polished thin sections were counted features as a function of feature size at a fixed magnification (20x). Applying the equation of the comparable technique of cord-length measurements on fracture surfaces

\[ N (L) = A L^{2-d} \quad (Krohn, 1988) \]  \hspace{1cm} (67)

where $N$ is the number of features per unit length and $L$ the length measured, one could obtain the fractal dimension of the rock-pore interface from the gradient of a log-log plot ($L$ plotted in a log-scale to base 2).

Because of the restricted time for use of the technique, and the preference in determination of the pore size distribution (see SEM) the features were only measured at the lowest magnification. The resulting weak “apparent” fractal dimension are therefore based on a limited range of data points. Moreover "overlapping" pores could distort the power law behaviour.
3. 2. 10 Scanning Electron Microscopy (SEM) - Analysis

The SEM is a valuable tool as it enables analysis at a specific spot of about 1 µm diameter on the surface of a specimen and photographs at very high magnification (from 20x up to 100000x) in three dimensions. For examination of fresh broken rough surfaces the SEM is operated by collecting low energy emitted electrons and high energy reflected electrons (reflection-emission mode). Examinations of polished surfaces are carried out in the reflection mode which collects only the backscattered electrons (BSE). During both of the operating modes the energy dispersive X-ray analysis system (EDX) for mineral identification was used.

Normal reflection-emission mode of operation:
The four samples (O6, G16, H1, H3) examined were chosen for their characteristic difference in permeability.
The aim was the generation of a 3 dimensional image of the specimen and the pore cast with a maximum of resolution and X-ray mineral identification.
For this purpose it was employed the EDX-analysis for element detection. From important feature were taken photographs.

Principle
The principle of SEM is to bombard the surface of a sample with an focused electron beam. Some electrons are reflected as backscattered electrons (BSE), whereas others cause the emission of low energy secondary electrons (SE). Both of them are collected by a scintillator and converted into high resolution image.

Limitation:
- The lightest element which can be detected is sodium (Z=11).
- Coating must not contain any element for which the analysis is required (carbon outside the detection range).
- Surface topography affects factors such as the intensity and depth of X-ray production and the absorption effect. Rough surfaces can also obstruct the path of X-rays to the collector and backscattered electrons and X-rays can produce radiation from areas outside the primary target area of the beam (fig. 17).

Preparation:
A portion of each sample was broken up and a representative fragment from fresh interior of the rock was mounted on a 12.5mm diameter aluminium stub using a glue. The surface should be as flat as possible to improve analytical work. After mounting the samples on the stubs, the sides of the samples were coated with carbon (conductive medium). Then the sample were coated with a 25 Å thick conductive gold layer to reduce the electrical charge which builds up
on the specimen surface due to electron bombardment. Unfortunately, it was found that with the thin gold coating, strong picture charge build-up effects caused picture resolution to be poor (bright spots and photographs have bright horizontal lines). On two porous samples it was therefore necessary to re-apply a gold coating in order to increase the thickness.

The casts samples are made by saturating the samples under vacuum for 4-5 h with a plastic resin (same was used to impregnate sample for thin sections); after drying the sandstone was dissolved with hydrofluoric acid (one day and one night).

Reflective back-scatter mode of operation
Polished sections of 14 samples were analysed. They include the entire H-series, G13, G15, G16-G19, and O6-O8.

The aim was to obtain the best image contrast between grains and pore space (and on one sample between quartz-grains, pore space and heavy minerals) and to identify minerals by X-ray analysis.

Principle and operations:
The back-scatter electron microscopy differs from the normal emission-reflection mode (SE-BSE-mode) in that way that the secondary electrons are prevented from reaching the collector. Thus the degree of contrast of an image is according to the minerals atomic number because the reflection coefficient increases as the atomic number increases and hence particles with higher atomic number appear brighter.

During operation in the normal reflection-emission mode this effect is usually masked by topographic effects, but can most effectively be utilised with a BSE detector to produce images of polished surfaces. The image is then thresholded, setting upper and lower limits to show all the grains up as white to grey and the pore space as black. A grid of points is then superimposed on the threshold image. The magnification (36x) was set on the lowest limit.

The other parameters were set as seen below:
The beam then scans this threshold image and stores the image. It is this stored image on which the computer conducts its analysis and calculations:

Parameter measured:
- Percentage of area of features of a certain pore size fraction
- Number of features of a certain pore size fraction

Parameter calculated:
- Mean pore diameter distribution (weighted by features detected and by area)
  It is the mean Feret diameter (Feret, 1931) calculated from 60 diameter projections around the feature area. The total number of grid units occupied by the feature is converted into square microns. This method appears to be more scientific than to assume a 3 dimensional spherical shape of the pores and then to calculate the diameter of a circle whose area is equal to the feature area. The diameter obtained would be smaller for non circular pore shapes, compared with the measured 2 dimensional mean Feret diameter.
- Maximum pore diameter distribution:
  The maximum diameter is expressed as the length of the line joining the touching points of the maximum and the minimum Feret diameter projection.
- 2D porosity

Preparation:
Polished thin sections of the samples impregnated with blue plastic resin. Then coated with carbon as it must be conductive.

3. 2. 11 Point Counting
The point counting under transmitted light was performed with the same 14 polished sections as already mentioned in the SEM chapter.

It was tried to determine the area of pore space, highlighted by a blue resin. In parallel it was tried to collected the percentage of cementation, separated from the detrital grain by a black dust line. The point counting was carried out on 400 points, seperated by 0.3 mm point space. The detrital grainsize of six samples (H1, H3, H5, O6, O7 and O8) were determined by the employing the method of Feret’s statistical diameter. The diameter is defined by two parallel
lines which are touching the outer grain borders. The parallel line lies perpendicular to the point counting transections (fig. 18). A maximal number of 120 counted points were examined to be accurate to represent the samples grain size distribution. It was chosen a point space of 0.3 mm. The diameters are grouped in logarithmic grain size fractions to the base two.

![Fig. 18: Determination of Feret's statistical grain size diameter by point counting](image)

### 3.2.12 Cold Cathodoluminescence

*Unlike much scientific equipment, it is justifiable to use this technique without good reason, simply to see what can be seen.* (Dr. Gordon Walkden)

Cathodoluminescence colours were generated of a polished thin section sample of the H-series and a contact metamorph cemented sample prepared by Munro (1997). The aim was to examine the silica overgrowth of the grains with special emphasis on colour and on any zonation.

**Principle**

Fluorescence is an effect of photon bombardment. In the case of cathodoluminescence the source by which the emission is produced, is an electron beam. This beam causes electrons to jump energy levels, and in substances there is a significant large energy gap to be negotiated between energy levels, the transition of an electron back to its ground state is accompanied by emission of energy in the form of a photon.

**Equipment and Operations**

The electrons are generated in vacuum conditions and under voltages of around 15 kV. at the cathode end of a tube, and many hit the specimens by the beam released at the hole in the anode. The instrument used is a Citl Cold Cathode Luminescence 8200 mk3 with a gun current of 620-630 mA, in combination with an ordinary light microscope. The specimen is housed in a chamber that is attached to the microscope stage. The chamber is equipped with lead class windows (x-ray generation possible!) at the top and the bottom to permit normal
petrographic observations. Because of the chamber and the demands of focusing, the equipment specific limit of objective lenses was x20. Photographs were taken using Fuji G-400 colour film with exposure times of up to 11 minutes. One should note that better results in the analyses of minerals with poor luminescence such as quartz were obtained by the use of hot cathode systems, which employ higher voltages (Bruhn and Bruckschen et al. 1996).

### 3.3 Whole Rock Analysis

#### 3.3.1 X-ray Analysis

X-ray fluorescence and diffraction analysis were performed on an unweathered and a weathered representative cemented material of the rock. In order to improve the detection of any different trace element concentration in cemented and uncemented formation, the > 20 µm size were also examined. The <2 µm fraction was only possible to measure in the highly cemented sample.

The qualitative X-ray diffraction analysis was used to obtain information on the content of minerals in the rock, in this case especially clay minerals. The charts were produced within a reflection range of 3-50° (2θ) by using a Philips 1730 Generator (CuKα radiation). The quantitative X-ray fluorescence analysis was carried out for major and trace elements on a ARL 8410 wavelength dispersive X-ray fluorescence spectrometer using a 3 kW Rhodium target end window X-ray tube.

**Principle**

The technique involves bombarding of the prepared sample with monochromatic X-rays generated from a X-ray tube (XRF: Rh-radiation, XRD: Cu-radiation).

X-ray are only reflected by the crystal lattice at angles meeting the Bragg equation:

\[
2d \sin \theta = n \lambda \tag{67}
\]

where

- \(d\) = spacing of the lattice (Å)
- \(\theta\) = angle of incidence
- \(\lambda\) = wavelength of radiation
- \(n\) = order of intensity

**X-ray Diffraction (XRD)**

During bombarding of a sample surface, which change its angle to the X-ray beam during the measurement in a distinct way, the angle and the velocity is measured in the diffractometer.

When the beam encounters a set of regular lattice planes at a specific angle of incidence then part of the beam is reflected at an angle equal to the angle of incidence. The monochromatic X-ray diffraction diagram shows the angles for 2θ (common minerals in
the range of 2-32°). Using Bragg’s equation one can then figure out the value of the characteristic spacing of the crystal lattice \(d\). This \(d\)-values identify the mineral in question.

Parameters:
- \(kV: 36\)  
- \(mA: 24\)  
- \(c.p.s.: 100\)  
- \(Filter: Nickel\)  
- \(Time constant: 10\)  
- \(Charts speed: 10 mm per minute\)  
- \(Scanning speed: 1 \text{ grad per minute}\)  
- \(Attenuation: 10\)

Radiation: Copper, wavelength: 1.54050

Limitations arise from the overlapping and similarity of reflections. Diffractometer accuracy is a function of the scanning speed, the chart speed and the accuracy of the indicated chart speed.

- **X-ray Fluorescence (XRF)**

  The result of bombarding a sample at fixed position is a polychromatic fluorescence X-ray spectrum, made up of the wavelength of each element in the sample. Afterwards the radiation is focused into a parallel beam and split into its component wavelengths by using a diffractometer crystal of known \(d\)-spacing. Because the \(d\)-spacing of the diffracting crystal is known, the angle through which a particular wavelength of radiation is diffracted can be calculated. The goniometer then moves to the detectors to the appropriate position for the particular wavelength of radiation which is characteristic of the particular element (fig.19). The intensity of this fluorescent radiation is proportional to the concentration of that element in the sample (concentrations of major elements in percent, concentrations of trace elements in ppm).

![Diagrammatic layout of the fluorescence spectrometer](image)
Sample preparation:
The first step was to dry the sample. Afterwards the “fly press” was applied to crush the rock. Then the crushed rock was powdered in an agate-mill for 20 minutes.

- XRD:
The measurement was done on an acetone smear of the powdered sample on a glass slide. The separation of the 2 < 20 µm fraction and the < 2 µm "clay"-fraction was done in a settling water column. The withdrawal times are taken from Galehouse (1971) under employment of Stoke’s Law for spherical quartz grains in a settling column of water.

- XRF:
The major elements were analysed on fused glass discs. For this purpose the rock powder was weighting out into small Pt-Rh-Au crucible in following proportions: 0.5950 gram of rock powder and 3 gram of spectroflux. The crucible was then melted at 1050°C for 30 minutes; and the melt was poured onto an aluminium plate held at 230°C. Then the disc was placed on a second plate held at 100°C and cooled over night. The trace elements were analysed on pressed powder pellets: About 10 g of the rock powder was put into the agate mortar and mixed with 6-7 drops of PVA to improve the binding qualities of the pellets. Then the sample was put into a specially constructed die. The die was put into the hydraulic press which exerting a pressure of 15 tons for one minute in order to receive well formed sample briquettes.
IV. Results

4. 1 Sedimentology

4. 1. 1 Composition

The composition of the minerals was examined with four different methods. The optical analysis with the microscope (petrography) and element analysis with the EDX-analyser (SEM) gave information on distinct minerals, whereas the XRD and XRF analysis provided information about the elemental composition of the whole rock, from which the mineral composition was inferred.

4. 1. 1. 1 Transmitted Light Petrography

Samples of different permeability were analysed: O6, G16, H1, H3.

All the samples consist almost exclusively of quartz (nearly 100 %). Therefore, they can be classified as quartz arenite accordingly to Williams et al. (1954). Overall, the sandstones showed good sorting (excepting O6 which was moderately to well sorted), with rounded and well-rounded detrital grains (again O6 was the exception, showing small angular and subrounded grains), and no matrix. The solely monocrystalline quartz minerals are characterised by unit extinction and abundant solid inclusions of mainly tourmaline. Apart from these observed conformities throughout the samples, the following differences were detected:

- Sample O6 contained opaque dark minerals consisting of iron oxide (magnetite?, goethite?, ilmenite?). They were found as a coating of quartz grains or as accumulations in the pore space (Fig. 20).

- Samples G16, and especially H1 and H3 showed development of quartz cement, which can be distinguished from the original grain by a thin dust rim (Figs. 21 & 22). The coating (dust rim) does not appear to have a negative effect on the thickness of the overgrowth (cement).

- Sample G16 contains small amounts of muscovite (Fig. 23) and an altered detrital clay pellet (Fig. 21). Muscovite was also detected macroscopically in other core plugs which were cemented or well compacted (G19, G15, H3 and O6). A more detailed description of the mineralogy of the Lochaline sandstone has been carried out by Humphries (1961).

- Sample G16 contained obvious in-filled burrows. In the hand specimen, burrows are characterised by an irregular, Y-shaped branching pattern and dark colours. They have a diameter of approximately 0.4 cm. In addition to G16, burrows were also present in core plugs G13, G15, G17 and perhaps in H4. The thin section revealed that burrows are lined with clay minerals, but that the filling shows no difference in composition to the
surrounding sediment (Fig. 24).

The high textural maturity characterises the super mature stage of the sandstone after Folk (1951). The lack of any unstable rock fragments points towards a high compositional maturity.

4. 1. 1. 2  SEM

EDX-analysis on broken surfaces:

Interesting features were chosen for EDX-analysis during the SEM analysis of the samples O6, G16, H1 and H3. Autigene illite was identified in samples G16 and H3 as a pore filling mineral (Fig. 25). TiAl-oxides were detected as relicts on grains (Fig. 26).

EDX-analysis on a polished sample (H6):

In the analysed representative sample the following minerals were found: aluminium oxide (Figs. 27 & 28), muscovite (Fig. 27), apatite (Fig. 28), rutile and anatase (Fig. 29). The mineral assemblage apatite-muscovite was observed three times.

Solid onlate inclusions of long shapes (30 µm to 40 µm) consisted of ilmenite, muscovite and frequently biotite, whereas more spherical inclusions (10 µm to 20 µm) were formed by sphene, epidote and zircon.

Examination of the dustline (also visible under transmitted light microscope) between the detrital grain and the quartz overgrowth by EDX-analysis revealed a pure SiO₂ composition. The surrounding quartz probably causes this apparently pure quartz composition. The dust line has been observed in other sandstones as mainly consisting of void space (Pittman, 1972) or of fluid inclusions (Morad et al., 1991). Nevertheless, the bulk of the dustlines were not recognisable in the backscatter SEM mode. This feature indicates the absence of significant variation in chemical composition due to the filling of the voids by impure secondary quartz and/or impurities like amorphous iron and aluminium oxides.

Distinction between quartz overgrowth and detrital quartz core by means of the BSE detector was not possible, and thus reflects the absence of significant variations in chemical composition.

SEM-microprobe element analysis of the overgrowth itself was performed at different distances from the detrital grain and confirms a pure SiO₂ composition at all positions. The amounts of trace elements were below the detection limit of the EDX-analyser. There was no visible zonation of the overgrowth in the backscatter mode of the SEM.

The feature analysis determined the area of very bright spots (=heavy metals) as 0.02%, which is consistent with the TiO₂-value obtained by XRF (see below, section 4.1.1.3).

The EDX-ray charts are collected in Appendix 8.4.
4. 1. 1. 3 XRD / XRF

X-ray Diffraction

XRD analyses were carried out on two H-series samples.

- H6 - Representing unweathered cemented sandstone
- H7 - Representing weathered cemented sandstone

The weathered sample was expected to contain a greater clay content due to weathering, which, it was hoped, would aid the detection of clay peaks. Additionally the < 2 µm grain fraction of cemented H-series and uncemented O-series samples were also submitted to XRD analysis (H6 & O9).

Observations at 2θ angles between 3° and 32°:

Peaks at angles characteristic for the crystal lattice spacing of quartz (d=3.34 Å and 4.26 Å) appeared in the diffractometer charts of both whole rock samples (Appendix 8.6) together with an unusual hump between 12° and 7°.

Amorphous aluminium oxides or iron oxides might be responsible for the above-mentioned hump. The chart did not show the characteristic peaks of allophane and imogolite, two common aluminium oxides. Furthermore, no aluminium oxide was detected in the XRF analyses (Appendix 8.7). Ferrihydrite would be a common iron oxide, but the white colour of the rock and the peak at larger angles than expected argues against its presence in the rock.

Another explanation for the hump could be reflections of a mix of clay minerals, which have characteristic crystal lattice spacing in the range of question. Further investigation with a fine rock fraction (< 2 µm) result in an insignificant hump between 5.5° and 6.5° in the H-series, which may indicate the presence of montmorillonite. Nevertheless, quartz still predominates (intense peaks at 26.6° and 20.8°). Analyses of the small sized “clay-fraction” of the O-series also reveal a predominance of quartz, but also minor peaks at 25.3° and 8.6°. The first could be explained by the previously described presence of microscopic diagenetic anatase (TiO₂). The latter peak could represent muscovite (with the second muscovite peak at 26.8° masked by the intense quartz peak).

All four XRD-charts reveal no evidence of any precipitated opal-A within the rock, which would be indicated by a diffuse hump between approximately 19° and 25°, according to Williams and Parks et al. (1985).
Observations at 2θ angles above 32°:

The unweathered sample (H6) showed peaks at the characteristic angles for quartz, however their intensity distributions did not coincide with the patterns usually associated with quartz. Contrasts in intensity distributions between weathered and unweathered sample (H7 respectively H6) were found at two peaks (corresponding to d = 1.82 Å and 2.46 Å), which were greatly increased in intensity in the weathered sample. A possible explanation for this might be the orientation of quartz grains, or more likely a preferential alteration of the crystal faces 11'22 and 11'20 representing the above-mentioned d-spacing. Also, the unweathered and weathered samples show a sharp, intense peak corresponding to a spacing of 2.24 Å indicating the presence of the uncommon quartz crystal face 11'21. This may be due to preferential growth of this particular crystal face. A minor peak occurs in the unweathered sample (H6) at 46.9°, which could not be identified with Chao’s 2θ table (Chao, 1969) for common minerals.

The XRD charts are shown in Appendix 8.6

X-ray Fluorescence

The experiments were carried out on the same weathered and unweathered samples of the H-series (see above). The 20 µm to 2 µm fraction was analysed for sample H6 and O9 (only trace elements). The less than 2 µm fraction was analysed only for the sample H6 since the sandstones of the O-series are coarser grained.

Major elements

A good major element analysis should have a total oxide weight percentage of 99.5 to 100.5. In this case analyses had an oxide weight percentage of 101.16 to 101.11, which is commonly observed in XRF for almost monomineralic quartz samples, and is due to problems in the analysis routine.

The analyses (Appendix 8.7) showed a predominant quartz content and a very low content of TiO (0.02%) after processing. The analysis of the weathered sample (H7) showed a slightly higher content of TiO₂ (0.04%) and the presence of P₂O₅ (0.01%). No iron was identified despite the presence of occasional iron staining on hand specimens.

In the raw data charts of the cemented samples (H6 & H7), a higher Fe₂O₃ content than TiO₂ was detected. The loss of iron during data processing is a problem caused by the unusually high silica content. One should note that Chatten et al. (1971) also recorded up to 0.47% Al-oxide and up to 0.35% Ca-oxide in the Lochaline Sandstone.
Trace elements

A. Comparison of weathered and clean cemented quartzite

Antimony is the most abundant element (29 ppm) in the clean cemented quartz (H6), followed by Cr (12 ppm), Pb (11.9 ppm), Ni (6.8 ppm), U (5 ppm) and Mo (0.1 ppm).

The element Rb, Sr, Nb, Zr, Y, Ba, Zn, As, Bi and Th were qualitatively detected, but were below the quantitative detection limit (2 ppm).

The content of Nb, Cr, U and Mo is slightly increased in the weathered sample (H7) (up to 2.3 ppm). The elements Ni, Pb and Sb were slightly leached out due to weathering (decrease in the range of 1.6 ppm to 2.4 ppm).

However, there exists no significant difference in the trace element pattern between the weathered and unweathered samples.

B. Comparison of clean cemented and uncemented quartzite

The element Sb is the most abundant element in both samples. The uncemented sample (O9) is enriched in Zr, Cr, Pb, Mo and Sb, but no significant difference exists in the Ni content.

The 2 to 20 µm and clay (< 2 µm) fractions of the samples showed both enrichment in all of the trace element apart from Sb, which could only be detected in traces (<2 ppm). In particular, the elements Ni and Ba are massively concentrated in the smaller grain size fractions (Fig. 30). The cemented sample (H6) is also Cr-enriched.

C. Comparison of LSst and Plateau Basalt data

The relative low content of Ni, V and Cr in the Lochaline sandstone, which are typically abundant in basalts (Taylor and Smith, 1975, Thompson, 1982), suggests that silica was probably not contributed from basalts during extrusion or weathering.

Only a comparison of the smaller grain size fraction of the LSst with the trace elements of the Plateau Basalt yielded useable patterns. Whereas the peaks of the elements Cr and Sr are not reflected in the LSst, the elements Ba and Ni shows similar peaks in the two formations (Fig. 30). The elements Rb, Nb and Y reveal the same low abundance in the basalt and the sandstone. However, similar trace element peaks are observed in pure quartz sands, which are not covered by basalt (Götze and Lewis, 1994). The XRF data table is given in Appendix 8.7.
Fig. 20: **Sample O6**: PPL & XPL micrographs showing grain fracturing (A) due to mechanical compaction reducing primary porosity. Note the moderately well rounded uncemented quartz grains, which are coated with iron oxide. Some grains are embayed (B) as a result of corrosion during diagenesis, which may lead to an enhancement of porosity.
Fig. 21: **Sample G16**: PPL & XPL micrographs illustrating pressure solution. One quartz grain has undergone solution leading to the penetration of one grain by another (concave-convex contacts). Note that the penetration occurs after cementation (A). The rounded to subhedral grains show syntaxial quartz overgrowth which reduces the intergranular porosity. Overgrowth has been formed after chemical solution marked by embayed detrital grains (B). Top right hand side: burrow wall picked out by clay patches (C). Top left hand side: partial dissolution of detrital clay pellet resulting in micro-porosity (D).
Fig. 22: **Sample H3:** PPL & XPL micrographs revealing a high degree of synaxial quartz overgrowth. The boundaries between detrital core and overgrowth are marked by distinct coatings of iron oxide on the original detrital grains. Secondary porosity occurs here due to the dissolution of material (fossil?) and due to rock fracturing, which extends across and between the grains. (Thin sections are impregnated with blue resin to show porosity).
Fig. 23: **Sample G16**: PPL & XPL micrographs illustrating the initial stage of syntaxial quartz overgrowth on monocrystalline quartz grains. The overgrowth shows rhombohedral (A) and prismic (B) faces. Dissolution of a detrital muscovite flake along the cleavage produced secondary micro-porosity and a residue of undissolved material close to the pore may be illite (C).
Fig. 24: **Sample G16:** PPL & XPL micrographs showing a longitudinal section of a burrow wall. There are no differences in composition and texture between the burrow filling (right hand side) and surrounding rock. A thin “skin” of brown clay minerals marks the burrow wall (A), and results in the formation of micro porosity which may lead to anisotropy in the fluid permeability. Top right hand side: Oversized pore space is a good indicator of secondary porosity due to dissolution of material, the shape suggests it to have been a microfossil (B).
Fig. 25: **Sample G16:** SEM photograph showing authigenic illite precipitated in a pore throat, indicated by the extensive development of interwinded plates, which produced micro porosity.

Fig. 26: **Sample G16:** SEM photograph showing hexagonal flakes of quartz which partially cover the quartz grain. The flakes tend to be aligned parallel to the surface (A). The quartz grain is also covered by irregular ridges (dissolution ridges?), composed of Al-Ti-Oxide after EDX-analysis (B).

Fig. 27: SEM photograph showing an altered, rounded apatite and aluminium oxide in a pore.
Fig. 28: SEM photograph illustrating muscovite exfoliating along its basal cleavage and developing micro-porosity. Growing apatite and quartz within the muscovite foliation causes additional splitting of the muscovite. The rounded particle close to the muscovite has a composition of pure aluminium oxide.

Fig. 29: SEM photograph revealing replacement of certain parts of the detrital grain. Sedimentary anatase TiO$_2$ (bright colour) has replaced components of the derived sediment grain, leaving quartzose portions unreplaced.
Fig. 30: XRF-analysis of H and O-series Lochaline sandstone: trace elements of the Lochaline Sandstone and the Morvern Plateau Basalts. For the latter, data from Thompson (1982) were used. The small grain size fraction and the basalts reveal similar peak concentrations of Ni and Ba. The trace elements are sorted with increasing atomic number.
4.1.2 Cathodoluminescence

Cathodoluminescence was employed to obtain additional information about the quartz overgrowths. The observed luminescence was stable under electron bombardment; nevertheless, the colours shown in the figures do not represent the true colours. The photograph of the grain and overgrowth in Fig. 31 is closest to the two distinctive CL-colours observed.

Overgrowths detrital grains generally do not luminescence (McBridge, 1989). However, in the case of the LSst, the quartz overgrowth contains widespread domains with reddish-brown CL together with some non-luminescing zones (Fig. 32). Similar zonation has been reported from the Fontainebleau Sandstone (Bruhn et al. 1996), in which the luminescing zones have higher Fe concentrations than the non-luminescing zones. In general, cathodoluminescence colours in quartz are attributed to the lattice order, stress state, Al concentration, and the occurrence of trace amounts of positively charged ions with small ionic radii (Sprunt, 1981 and Matter and Ramseyer, 1985).

The position of the dustline observed under transmitted light microscopy corresponds very well to the change in CL colour. The reddish-brown CL colour of the overgrown quartz is typical of relatively defect-free growth of quartz at low temperatures (Marshall, 1988) and is likely to arise from differences in trace elements abundances, which in turn may reflect variations in redox conditions within a shallow freshwater aquifer. The similarity between CL colours and styles observed for LSst and Fontainebleau sandstone is consistent with such a hypothesis.

It should be noted that examination of contact metamorphic cemented samples of the Lochaline sandstone, prepared by Munro (1997), revealed cathodoluminescence colours which are identical to the colours of quartz overgrowths in the G- and H-series.

Sometimes the CL image allows distinction between fracture healing (dark blue) and detrital (light blue) quartz types (Fig. 31).

Despite the fact that the provenance studies are beyond the scope of this thesis, one should note that the detrital grains show two distinctive patterns of cathodoluminescence (Fig. 33):

- Light blue luminescent detrital grains (38% occurrence) This is characteristic of an igneous origin
- Dark blue to brown luminescent detrital grains (62% occurrence).

The photographs reveal a far better resolution than the transmitted light photographs. CL enables clearer identification of grain surfaces and pore shapes. The cathodoluminescence technique also detects emissions of grains that lie a few microns behind apparent pores detected under transmitted light (Fig. 34)
Fig. 31: PPL and CL micrographs show an often observed feature in LSst: Highly luminescing bright edges of grains and fracture healing. Fractures are marked by darker blue CL colours and occasionally through impurities that are visible under PPL. It should be noted that the colours in this CL micrograph are closest to the observed CL colours.
Fig. 32: PPL and CL micrographs reveal that the area enclosed by the dustline shows differing cathodoluminescence colours than the surrounding overgrowth area. A change in colour from blue to reddish-brown is observed. The wavy overgrowth contact between two grains can be interpreted as first signs of pressure solution (A). The brown patches in the PPL micrograph is “burned” blue resin. Concentric non-luminescing and luminescing zones are observed within the overgrowth (B).
Fig. 33: PPL and CL micrograph shows the two different CL-colours of the detrital grains and the uniform colour of the yellow-reddish overgrowth cement.

Fig. 34: XPL micrograph showing the same colours of grains and overgrowth, which indicates that the axis of the overgrowth has the same orientation as the one of the grain. The CL micrograph shows an atypical cement, which does not grow on grain surfaces (B), and it shows more grains than detected by XPL (A).
4.1.3 Grain size

The grain size was measured employing two methods; (i) by point counting thin sections under transmitted light, and (ii) by employing a relationship with the mercury injection capillary pressure (MICP) data (Eq. 6a in section ???).

Point Counting

The grain size determined by point counting was carried out on three samples of the O-series and three samples of the highly cemented H-series. In order to obtain the original size distribution before cementation, only the detrital grain size was counted. Grains were detected in fractions ranging from 20-30 µm up to 600-800 µm. There exists no significant difference in the dominant grain size. In all samples, the arithmetic mean grain size lies within the 200-300 µm fraction (Fig. 42). This agrees with the average grain size of 240 µm for the LSst obtained from sieve analysis (Humphries, 1961). However, the samples of the O-series show a higher percentage of smaller grains than the samples of the H-series.

For one cemented sample (G16) the entire grain diameter (detrital grain plus overgrowth) distribution was measured by point counting. The resulting median diameter of 259 µm shows that the grain diameter does not change significantly with cementation. However, the cementation resulting from this small increase in grain size can produce decreases of ??% in porosity and ??% in permeability (see section ???).

![Detrital Grainsize Sum Curve](image)

Fig. 42: Grain size distribution determined by point counting (??? points).

Capillary Pressure Data

The grain size distribution was calculated from the MICP data of 21 samples of the H-, G-
and O-series. Grains up to a size of 1600 µm in diameter were detected. The arithmetic mean grain size decreases with increasing cementation (O-series: 730-920 µm; G-series: 300-700 µm; H-series: 140-420 µm). The samples of the H- and G- series reveal a broader grain size distribution in comparison to the peak-like O-series. The grain size sum curves of the O-series are more uniform than the curves of the other two series (Fig. 43). Nevertheless, the absolute apparent grain sizes obtained from this method appear not to be representative of the real grain sizes of the samples. Almost all samples show significantly larger MICP-derived mean grain sizes than those obtained by point counting (see above) and by sieve analysis (Humphries, 1961).
Fig. 43: Grain size distributions calculated from mercury injection capillary pressure data.
4. 1. 4 Thickness

The base of the sandstone outcrop is just above sea level near Lochaline and climbs north-westwards up to 360 m at Beinn na h-Uamha (Fig. ??). In the mine at Loch Aline, between three and eight metres of sandstone are exposed. At Beinn na h-Uamha similarly thick sequences of up to 8.9 metres are observed. However, neither the top nor the bottom of the LSst sequences is exposed at Beinn na h-Uamha. Therefore, the logs in Fig. 45 are giving the minimal thickness of the formation at this location.

At the south-eastern side of Beinn Iadain, only minor outcrops are exposed, and no exposures were found around Beinn Ithearlan during a field mapping project. Along the eastern shore of Loch Aline the sandstone is approximately 12 metres thick (Lowden et al., 1992). The sandstone also attains progressively higher elevations eastwards, and has only six metres thickness about 1 km east of the Loch (Chatten et al., 1971).

The variations in thickness may be caused by pre-Tertiary basalt extrusion erosion, rather than by deepening of the basin southwestwards. Nevertheless, a decrease in bulk grain size westwards between Loch Aline and Gibrun at Mull (Bailey, 1924) could indicate a deepening of the basin westwards.

4. 1. 5 Architecture

The following description is refers exclusively to the outcrops of the LSst at Beinn na h-Uamha (Fig. 44). Despite the lack of sedimentary features, the LSst was separated into four units (Fig. 45), which is mainly based on changes in grain size:

A massive sandstone facies (D), up to 3 metres thick, forms the base of the sequence. At outcrop one, three and five a coarsening-upward from fine to medium sand can be observed. The following horizon (C) is characterised by thin parallel layers of very fine sand to fine sand, in strong contrast to the lower massive sandstone (D). The C horizon (20 to 30 cm thick) is occasionally separated from the lower massive sandstone by a thin black layer. This layer may include clay-grade organic material or heavy minerals and is suggested to be an isochrone. The next unit (B) again consists again of a massive homogeneous fine to medium grained sandstone of several metres thickness. A gradual increase in grain size to pebble-size marks the uppermost part of the exposed sequences which are up to 90 cm thick. This horizon was defined as the fourth unit (A) and was chosen as a "marker" horizon because of its lateral continuity. However, one should be aware that the base of the unit is probably erosive and therefore not necessarily an isochrone.
4.1.5.1 Structures and Interpretation

Unit A:

Description: This unit consists of a thin coarsening-upward sequence of poorly sorted pebble-rich sandstone in a medium to coarse grained matrix sand. Rounded to angular pebbles make up the rock. The pebble amount increases gradually upwards constituting up to 25% of the bulk rock. The pebble size averages 0.6 cm but occasionally up to 1.5 cm large polymorph quartz pebbles were found. Sedimentary structures are very rare. No contact at the top of the sequence can be observed since detritus and grassland cover the 4 to 6 metres thick gap to the overlying massive basalt exposure. The contact to underlying unit B is gradual.

Special features:
Outcrop 1: Vertical quartz veins, 1.5 cm thick coal seam.
Outcrop 2: A 5 cm thick, burrowed horizon is highlighted by iron precipitation at the top of the unit; lenticular accumulations of coarse sands (pocket-like) which are laterally developed within fissures in the upper part of unit B.
Outcrop 3: Silicified white structures (shell layer?); two layer of caved horizons (thickness 5 to 10 cm); parallel lamination.
Outcrop 4: Vertical "pockets" of coarse grained sand.
Outcrop 5: Iron oxide nodules.

Interpretation:
The pebbly sandstone may represent wave-reworked sands. It could be related to wave processes concentrated on a bar crest.

Unit B:

Description: This massive unit is composed of fine to medium grained sandstone. Occasionally, small areas of cross bedding are visible, but they are rare and non-persistent. Walled burrows (Ophiomorpha?) are marked by iron precipitation. The burrows are predominantly vertical, up to 1.5 cm in diameter, and up to 10 cm in length. Cemented horizons are seen exclusively in this unit. They range from 5 cm up to 125 cm in thickness. Iron staining is absent. Columnar structures are observed within the cemented as well as within the uncemented units. The base of the thicker horizon is wavy (erosive?) and a large variation in thickness can be observed laterally.

Special features:
Outcrop 1: Amalgamation surface; small lenticular thin quartz cementation bands; trough cross bedding.
Outcrop 2: Yellow-red small patches of iron precipitation; lateral increase in amount of pebbles; thin layer with 6 to 20 mm pebbles in diameter, vertical and horizontal fractures.
Outcrop 3: Parallel cross-bedding with varying angles in north to north-west direction; trough cross-bedding; low angle cross-bedding; amalgamation surface (dip: 11° north-wards); synsedimentary deformation structures (slumping?); vertical patches of massive iron precipitation.

Outcrop 4: Parallel cross-bedding with varying angles.

Outcrop 5: Interlayer of unusual black-brown (ferruginous) coloured sand which contains iron rich spots between two highly cemented horizons. Two amalgamation surfaces which are cutting-off lower cross-bedding; hummocky cross-bedding; low angle and trough cross-bedding.

Interpretation: It is thought that this unit had tabular cross-bedding but that post-depositional bioturbation has largely destroyed this primary fabric and homogenised the sediment. The lack of glauconite and fine particles in comparison to the stratigraphically lower Greensand has been interpreted as the result of high energy reworking processes (Humphries, 1961). The walled burrows support the high energy environment, shallow water interpretation. The lack of shell material should not be seen as a primary factor but as a result of decalcification. Decalcification is suggested by Lowden et al. (1992) "from the absence of calcareous fossils in the LSst which otherwise has abundant evidence of faunal activity".

The coarsening upwards indicated by a gradual increase in the amount of pebbles between Units A and B and the regional setting suggest these sediments to be deposits of a sandbar in a wave-dominated delta front.

Unit C:

Description: The moderately sorted, fine sand unit is characterised by horizontal or sub-horizontal parallel lamination and, in some areas, by parallel tabular cross-bedding in the basal part. This feature is only visible when seen in an exposure trending parallel to flow-direction, where the foresets are seen in longitudinal section. Foreset heights average 10 to 15 cm, and foreset dip ranges from 35° – 20°. Because there is little visible grain size or mineralogical variation, the lamination is not easy to observe. Despite the parallel laminated units dipping with a low angle, the lamination appears to remain parallel to the bed surface. A two mm thick, laterally discontinuous dark layer is occasionally visible at the top and the bottom of the unit. This layer is composed of fine organic material or heavy minerals. The contact to the lower unit D appears to be erosional.

Special features:
Outcrop 1: Dark (current?) ripples, 2 to 3 mm thickness in the ripple troughs and with approximately 3 cm wavelength.
Outcrop 3: Westwards thinning out of the unit.
**Interpretation:** Planar, parallel laminated sandstones are typical of high energy, shallow water conditions, and are therefore characteristic of beach foreshore deposits. The scale of the type of the cross-bedding is characteristic of small-scale sandwaves, which are formed usually under moderate energy flow conditions.

**Unit D:**

**Description:** This unit is similar to Unit B but is characterised by sedimentary structures highlighted by iron oxide precipitation. Horizontal structures, such as amalgamation surfaces and low angle cross-bedding, or parallel lamination in the lower part of the sequence, dominate. Bioturbated inhomogeneous coloured sediment marked by patches of iron oxide increase towards the top of this unit. The unit comprises a coarsening upward sequence from very fine through fine to medium sands.

**Special features:**

- **Outcrop 1:** Unit separated into a white coloured upper part, and a light brown lower part; trough cross-bedding; parallel or low angle cross-bedding in the upper part; vertical fractures and patches of iron oxides in the lower part.

- **Outcrop 3:** Discontinuous parallel lamination (15 cm thick); horizon of yellow flakes (?mud flakes); lenticular iron oxide patches; isolated iron oxide nodules; low angle cross-bedding visible by small variation in grain size; 2 mm to 3 mm thick layer of coarse grains (0.5 mm); muscovite layer; trough structures; horizons highlighted by truncated horizontal burrows; hummocky cross-bedding.

- **Outcrop 4:** Horizon with yellow flakes; single iron oxide nodules; parallel cross-bedding; soft sediment deformation (?water-escape structures).

- **Outcrop 5:** Iron oxide patches in the top 20 cm to 30 cm (?bioturbation) and dark minerals; parallel layers of 1mm to 2 mm thickness highlighted by iron oxide precipitation; parallel cross bedding; iron oxide patches up to 2 cm; horizontal flat structures; vertical fractures highlighted by iron oxide precipitation (5 mm thick).

**Interpretation:** The amalgamation surfaces, trough cross-bedding and hummocky cross-bedding could represent a lower shore face facies. The upward increase in grain size could indicate a transition from off-shore facies to shore facies. The intensely bioturbated horizon, marked by patches of iron oxide at the top of the unit could be interpreted as a result of declining sand deposition.
4.1.5.2 Trends

Vertical Trends

A coarsening upwards sequence from fine to coarse sand can be observed at Beinn na h-Uamha as well as in and around the Lochaline sand mine. Within the mine the trend was concluded from an increase in permeability (Chapter 4.3.2). At Beinn na h-Uamha the coarsening-upward units vary in thickness; in most places representing less than a metre of the overall sequence.
Bioturbation is present throughout Unit B but also concentrated in the top few centimetres of Units B and D.

At Beinn na h-Uamha, the colour of the sand changes from light brown at the bottom of the sequence to white at the top.

**Lateral Trends**

All four main units are traceable laterally over several hundred metres as seen in Fig. 45. This lateral continuity of units is in agreement with observations in and around the Lochaline sand mine (Lowden et al., 1992) where individual cross-bedding units can be traced over distances of tens of metres. Lowden et al. (1992) postulated a lateral extension of the units of the order of 100 metres from the observed constant dip and thickness of the cross-bedding units.

Unit B shows a strong variation in thickness with the thinnest sequence (1.6 m) on the eastern side of the profile (outcrop 4) and the thickest. Given the parallel laminated Unit C as an isochrone horizon, the remarkably thick Unit A in outcrop 4 could be linked to the thinning of Unit B. The parallel laminated sandstone Unit C thins out, but can be traced laterally over hundreds of metres without any significant change. Columnar (subsurface or subaerial?) weathering structures are prominent at exposures at the West Side of Beinn na h-Uamha.

Cemented horizons show lateral extension of no more than 10 metres. They occur as small few centimetres thick lenticular concretions, wavy bands, or as thick (up to 1.25 m) distinctive horizons. No lateral continuity between the outcrops was recognised, but the occurrence of cementation is restricted to Unit B. This observation differs from the laterally continuous two distinctive cemented horizons in the Lochaline sand mine. Cemented horizons are not linked to the existence of dykes in the neighbourhood.

In Unit A the pebbles size decreases westwards from 5-6 mm to 3-4 mm in diameter, as well as a decrease in the percentage of pebbles from 40% to 20% in the westward direction.

**4. 1. 5. 3 Geometries**

In the following each of the five examined outcrops is presented as a photograph and in a graphic format with special emphasis on the lateral development of the units and in particular of the cemented horizons. Vertical lines mark the position of the logs in Fig. 45.
Fig.46: (a) and (b); Photograph and graphical description of outcrop one. Outcrop one reveals at least two horizons with horizontal or low angle beds of highly cemented sand. The cemented layers are up to 30 cm thick and show a protruding nodular texture (c). Unit B appears to be massive, and shows a prominent development of vertical rills (c). One should note that only unit B shows this feature. In the upper and lower part of the sequence, the rills end in grooved “ring” structures (see also outcrop 2). Therefore, it is more likely that internal columns of the sandbody (of unknown origin) produce the rills rather than just weathering.
Fig. 47: (a) and (b); Photograph and graphical description of outcrop two. This outcrop revealed the thickest cemented horizon, with a thickness of 125 cm. The base of the horizon is uneven and the thickness decreases south-eastwards. Unusual ring-structures are the dominant feature within unit B (C). It appears that these structures are the “cut-offs” of vertical columns. Occasionally, vertical pockets are observed within the upper part of the cemented horizon.

Biogenic structures (burrows?), highlighted by iron precipitation, are visible at the top surface of the outcrop (d).
Fig. 48: (a) and (b); Photograph and graphic showing outcrop three. Unit C thins out towards the left-hand side. Further features are the cave in unit D, and the vertical patches of iron precipitation in unit B. Enlargements show the base of unit C and parallel lamination which cuts the lower cross bedding in unit D (c). A thin (organic?) black horizon between the units is also visible. Several walled burrows (*Ophiomorpha*) can be observed in unit B (d). Iron staining is linked to sedimentary structures as proven by truncated burrows (c). Low angle and horizontal structures are characteristic for unit D and marked by iron precipitation (f).
Fig. 49: (a) and (b); Photograph and graphic showing outcrop four. Unit A reveals the thickest sequence of all outcrops with 145 cm. Enlargement (c) shows iron precipitation along bedding planes. These are irregular in dip or curved and not laterally extensive by contrast to the horizontal bedding planes in unit C of outcrop four. Enlargement (d) shows a small seam of coal, which marks the contact between unit B and C. An increase in small black particles (coal?) towards the top of unit C is also observed.
Fig. 50: Photomosaic and graphic showing outcrop five. The annotated rectangles in the graphic mark enlarged detailed views, given as Fig. 51a to 51c

The basalt dyke has no influence on the slope of the strata – as may be suggested from the photograph which gives a slightly distorted the right view. One should note the variation in thickness of the cemented layers in Unit B. The thickness of the layers varies from 15 to 90 cm. The lower horizon wedges-out westwards, whereas the upper horizon thins towards the east. The base of the lower horizon appears to be horizontal.

The layer in between the cemented horizons shows an unusual dark-brown colour and contains iron-rich patches (Fig. 51a). The holes in the upper and lower cemented layer are interpreted as weathering features, with no differences in grain size were observed between the holes and the surrounding sand.

The upper part of Unit B develops rill erosion, but it is less pronounced than in outcrops one and two (Fig. 51b). In the middle of the outcrop a thin dark seam within Unit B (coal?) is present.

The planar cross bedding of Unit C can be seen in a magnification of section C (Fig. 51c). The upper left-hand part of the photograph shows trough cross bedding in Unit B. Iron staining and iron precipitation along fractures are prominent.
Fig.: 50a  Fig.: 50b

Fig.: 50c
4.1.6 Diagenesis

Three different methods were employed on the samples O6, G16, H1 and H3 to investigate the diagenesis in the LSst. There were (i) examinations of thin sections by transmitted light, (ii) cathodoluminescence and (iii) scanning electron microscopy. The results of the XRD/XRF element analysis and observations were also need to provide additional information concerning diagenesis.

Sample G16, H3 and H1 showed predominant overgrowth of quartz (silicification) and solution features.

Sample O6 showed effects due to compaction and solution, but no quartz cementation.

The processes of postdepositional change are relative simple but vary within the sandstone sequence. The examinations revealed two basic processes of importance for modifying the primary porosity in the sandstone:

- Leaching and silicification due to meteoric water influence
- Mechanical compaction and pressure solution (chemical compaction)

The very localised contact metamorphic effects adjacent to dykes are beyond the scope of this report, and examined in detail by Munro (1997). Iron staining is only seen as an insignificant process in changing the porosity of the formation.

Early diagenetic processes (leaching and silicification)

The already compositionally mature sand has probably been leached further by acidic meteoric water, removing all but the most resistant minerals (muscovite and quartz) and enhancing porosity over the entire sequence. The leaching process is evidenced by moulds of leached macrofossils in hand specimens (bivalve shells?), moulds of microfossils in thin sections (Figs. 23 and 28), oversized pores, and embayments on detrital grains (Fig. 58).

The leaching process could not be dated exactly, but probably occurs before silicification, and was intensive. This conclusion can be drawn from detrital grains with embayments, which show additional quartz overgrowths (Fig. 22), and from the only rare appearance of silicificated microfossils. One should note that the underlying Greensand is rich in calcereous microfossils. Silicified microfossils (Fig. 53), moulds of fossils in the highly cemented horizon, and embayments in the overgrowth cement of grains (Fig. 57) indicate that the two processes of leaching and silicification could have occurred together for a short time.

It is certain that the silicification occurred pre-burial since the detrital grains were in a loose packing without any significant compaction features (Figs. 23 and 54), with later cement occluding the original intergranular volume. The early stage of cementation is also confirmed
by a crossplot of the percentage of intergranular volume against intergranular cement (Fig. 52). Hereby, the uncemented O-series shows a higher degree of compaction than the cemented G- and H-series. Examination of the boundary between cemented and uncemented horizon in a thin section revealed a feature which may be interpreted as an erosion surface; viz cemented grains are showing abrasive cuttings (Fig. 55). This feature again suggests an early, near-surface precipitation of quartz. However, no reworked cemented particles were found.

XRF analysis shows a high content of Ni and Cr, which are typically abundant in basalts (Fig. 32). Hence one could suggest that silicia was derived from basalts during extrusion or subsequent weathering. For example, Judd (1878) has associated features within the sandstone with the overlying basalt: “The surfaces of sandstone over which the lava has flowed have been broken up into hexagonal columns by the contraction of the mass in consequence of its being heated and desiccated”. However, the peaks of Ni and Cr occur in both the cemented and uncemented samples. From the petrographic and stratigraphic point of view, the cementation horizons reveal a more complex history than can be accounted from a single thermal event for the following reason:

- Two modes of diagenetic quartz occur. There are predominant synaxial overgrowths and aggregates of euhedral crystals (Fig. 59). These types of overgrowth are not typical for high temperature precipitation. Amorphous quartz was seen neither in the thin section nor detected as peaks in the XRD-charts. Only an insignificant amount of “floating” quartz cement was recorded, and appears to be unrelated to the grain surface (Fig. 41).
- The zonation of some quartz overgrowths observed under cathodoluminescence suggests that the pore fluid had varying compositions during precipitation. Therefore, the silicification is presumed to be precipitated by a succession of events rather than by a single event.
- At different vertical levels several horizontal lenses of cemented sandstone were observed, which cannot be explained by a single thermal event.

Therefore, no particular time can be determined for the cementation. No evidence of the basalt/silcrete association was found, nor was any sign of direct or indirect influence of the basalts found (e.g. that the basalt could have act as caprock over the quartzose sediments providing a confined aquifer within the sands).

**Late diagenetic processes (compaction and pressure solution)**

Muscovite squeezed between more resistant quartz grains (Figs. 24 and 42) indicate a high final degree of compaction. Fracture and cleavage of brittle quartz grains resulted in tighter packing arrangements because framework-supporting minerals had been leached out (Fig. 20). Fracturing is, in most cases, not restricted to the detrital grain, occurring also in the overgrowths in the G- and H-series samples (Fig. 23). This indicates that the compaction
stage has to be dated mainly as a post-cementation effect. There are only a few concave-convex detrital grain contacts, indicating the insignificant role of pressure solution as source of the silica (Fig. 57). However, one should note that concave-convex contacts are also observed between grains with overgrowth (Fig. 38), indicating that the pressure solution affects the sandstone at a post-cementation stage.

At outcrops, iron precipitation is present in fractures, and also appears to follow also sedimentary structures. This suggests that the iron was precipitated from iron-rich ground waters, as a relatively recent event. The iron staining, observed within the mine and at Beinn na h-Uamha, is probably a still ongoing process.

Contact metamorphic alteration occurred after basalt flow, since the dykes cut through the plateau basalts.

Caves and dissolution features, which follow fracture planes, are probably a recent surface related weathering process and not included in the Fig. 51.

Fig. 51: Framework for a simple diagenetic history after deposition.

Fig. 52: Plot of intergranular volume versus intergranular mineral cement of the Lochaline sandstone, showing the amount of original porosity destroyed by compaction and cementation (diagram modified after Houseknecht, 1978). The samples of the G- and H-series have lost approximately equal volumes of porosity.
by compaction and cementation. The total number of samples plotted equals 13. The original porosity of the sand was assumed to be 40 percent.
Fig. 53: **Sample G16:** PPL & XPL micrographs showing a rounded conical shape, filled by syntaxial quartz. The shape suggests it to have been a microfossil, with secondary cement from early diagenesis.
Fig. 54: **Sample H1:** Good examples of planar euhedral crystal faces can be seen on these PPL & XPL micrographs, which have grown into pore space and so strongly reduced intergranular porosity. The cement thickness varies around the pore space and is therefore partially a function of crystal orientation and subsequent quartz overgrowth. Areas of quartz outlined by iron oxide or titanium oxide are interpreted to be the original grains.
Fig. 55: Photograph showing the enlarged contact between cemented (lower part) and uncemented (top part) horizons. The grain cut-off at top of the cemented horizon is thought to record an erosive surface. The uncemented horizon shows an unusually high percentage of small particles, which are probably reworked particles from the underlying horizon.

Fig. 56: **Sample O6:** SEM photograph revealing that the points of contact between quartz grains are sites of pressure solution. These appear as circular spots marking the contact between visible grains and formerly adjacent grains (removed during preparation).
Fig. 57: **Sample H3**: SEM photograph showing large corrosion embayments in a quartz overgrowth.
Fig. 58: **Sample O6**: PPL & XPL micrographs showing angular - subrounded smaller grains to rounded larger grains in a tight packing, moderately sorted. Note the embayed shape of some grains, which indicate chemical solution. Many types of grain to grain contacts exist: point, interpenetration, and most abundantly planar grain to grain contact.

Fig. 59: **Sample H3**: SEM photograph illustrating how quartz overgrowths with well defined crystal faces grow, meet adjacent overgrowths, and become a single crystal mass.
4.2 Porosity

The connected three-dimensional porosities were measured by fluid imbibition and gas expansion methods. The two-dimensional measurement of the total porosity was carried out via SEM image analysis and optical light microscopy by point counting. One should note that most of the techniques used differ not only in the parameters measured but also in the scale of investigation (see Fig. 6).

4.2.1 Three-Dimensional Porosity

The porosities derived from the calliper, helium, archimedes and mercury method are summarised in Fig. 60. In each single method similar porosity trends were observed. The highest porosity values were found in the O-series (~20 %), intermediate values in the G-series (between 6 and 13 %) and lowest values in the H-series (between 4 and 7 %). Comparisons between the methods revealed increasing porosity values in the following order: lowest values were derived from the calliper method, higher values from the Archimedes method, and the highest values were obtained from the helium method. This is due to the increasing accuracy and penetration of the methods (see below). The mercury method resulted in higher porosity values than the helium method for the O-series, but this observation cannot be extended to the H- and G-series. The higher porosity values with the mercury method for the O-series can be partly explained by the different surface structure of the samples; the rough sample surface (caused by the brittle grain structure) might not have been completely covered with mercury which would lead to a higher estimate of the bulk volume.

The calculation of the grain densities (grain density = bulk density * bulk volume/grain volume) can give an indication whether the pore or grain volumes have been determined accurately. Since the composition of the grains is known to be quartz, the densities should be close to 2.647 g/cm³. The calculated grain densities of the analysed samples can be seen in Fig 60. Grain densities obtained by the mercury-method were nearly always too high, the exception being H3 and G13. The lower value of H3 was caused by a low saturation of the pores (lower detected pore volume) due to incorrect pressure during injection (1 bar). The sample G13 revealed an exact density of 2.647 g/cm³ despite the fact that the measurement was carried out with a volume of mercury filling that was too large. This might indicate that
the mark on the dilatometer, which was renewed during the measurements, was positioned too low (in comparison to the calibrated mercury volume). This mis-marking could have led to the overestimation of the bulk density, which in turn would result in an overestimation of the porosity.

The helium method generated grain density values with the closest proximity to 2.647 g/cm². Therefore, it is assumed to be the most accurate method. Nevertheless, values tend to be slightly lower than the expected value. The reason for this may lie in unconnected pore spaces which cannot be reached by the helium gas, and which would be incorrectly counted as grain volume. Because the absolute pore volume is very low in the low porosity samples, the relative amount of unconnected pore space may be considerable.

The calliper method proved to be very poor due to inaccurate determination of the bulk volume. Due to the improvements in accuracy, the helium porosity calculated with a bulk volume determined by the Archimedes technique is 1 to 2 percentage points lower in the H- and G-series, and 5 to 6 percentage points lower in the O-series. In a few samples (H5, G20 and G21) the bulk volume was calculated as an average of the calliper and Archimedes bulk volume. This was done because the bulk volume measured with the Archimedes method was too low due to a insufficient core-surface dry-up, which has probably influence the wet weight, and also because the calliper bulk volume was too high due to surface vugs.

Porosities determined from all methods were plotted relative to the helium/Archimedes porosity determined for each sample to assess the degree of scatter in the porosity determinations from each method (Fig. 61). It should be noted that the choice of the helium/Archimedes technique as the standard was taken on the expectation that this combination of techniques produces the truest porosity. Over the entire range of porosities, the mercury porosity reveals high deviations of up to 50 %. The calliper and Archimedes methods tend to slightly underestimate porosity of up to 10 %, with an increasing scatter in the low porosity range (below ~ 7 %).

### 4.2.2 Two Dimensional Porosity

The analysis was carried out on a restricted number of samples (O6, O7, O8, G13, G15, G16, G17, G18, G19 H1, H2, H3, H4, H5), and used the same polished sections as used for SEM backscatter image analysis and for point counting carried with an optical light microscope.

The SEM feature analysis program enabled determination of the area occupied by pore space, which appears black in the photographs of Appendix 8.3.
The two-dimensional porosities obtained in this way should not be expected to be identical with the volume-based porosity, since it is not yet proven that the LSst is isotropic, and this type of porosity determination only agrees with volume-based porosity values for highly isotropic porosity distributions.

A comparison of the point counting porosity with the core porosity revealed a trend to overestimation, with single values up to 80% higher. SEM image analysis shows a general trend of underestimating porosity, up to 50%. One should see those results only as a rough trend, since the limited number of samples is statistically not stable.

The correlation coefficient of a poroperm trendline incorporating the two dimensional porosities are significantly lower than the correlation coefficients from poroperm plots incorporating porosities determined by the helium/Archimedes technique.
Fig. 60: Bar chart of helium, Archimedes, calliper and mercury porosities for all samples. Grain densities are given in the lower chart as an indication of the accuracy of the methods.
Fig. 61: Relative 3D and 2D porosities, referenced to 100% helium porosity.
4.2.3 Controls on Porosity

Since the helium porosimetry appeared to be much more accurate than all the other methods, the following discussion is mainly based on helium porosity.

It is important to define the mechanisms and structures which control porosity because porosity will profoundly influence all other petrophysical parameters presented in this study. For this purpose the samples, which are grouped in high porosities (O-series), intermediate porosities (G-series) and low porosities (H-series) are classified by types of porosity based on semi-quantitative observations of thin sections and SEM analysis. It should be noted that due to cementation any dissolved particles were easily to identify in the G- and H-series. However, in the uncemented O-series it was more difficult to distinguish between intergranular and any dissolution porosity. A qualitative distinction between micro-porosity, on the one hand, and macro-porosity on the other hand was obtained by employing Pittman’s definition (1979) to the data obtained from capillary pressure curves. Micro-porosity is defined as pores with pore apertures of less than 0.5 µm. The three porosity types (intergranular, dissolution and micro porosity), can be represented as end members of a ternary diagram that represents the total porosity (Fig. 62). The diagram gives no indication about the intensities of each of the processes leading to the different porosity types, presenting only the relative influence of the different porosities on the absolute porosity.
Primary Controls

In general the most important textural parameters in controlling porosity are grain size, sorting, shape, roundness and packing. All of these properties, apart from packing, are measurable and have been measured intensively for the porous part of the formation (Humphries, 1961). Grain size appears to be constant in the LSst and the good sorting changes little (Chapter 4.1.3). Sands with high sphericity and high roundness pack with minimum pore space. Therefore, it is expected that as sphericity and roundness decrease, porosity increases as a result of the bridging of pores and looser packing. However, in this study the changes of grain shape and roundness are caused by different degrees of cementation, and are therefore neglected, since they have no direct influence on the packing.

The isolated clay-lined burrows detected in some of the core plugs are not expected to affect the bulk effective porosity.

Secondary controls

Diagenetic processes are the main cause of the modification of porosity in this sandstone, and compaction and cementation are the main controlling factors.

The variability in the porosity of the highly porous part of the formation (O-series) could be caused by several processes during compaction. These are grain slippage, rotation and deformation. Van Engelhart (1960) suggested that grain rearrangement could reduce the porosity of sand from 40% to 28%. This estimation is very close to detailed calculations of Wilson and McBridge (1988) for Pliocene sandstone. In additional, authigenic quartz cementation as detrital grain overgrowth could be taken into account for the decrease in porosity in the cemented samples (series G and H).

In order to quantify the relevant influence of both factors, one has to define an original porosity. A model of uniform spheres in orthorhombic packing, which is assumed to represent the original configuration was assumed. Experimental studies result in porosity values of approximately 40% for this configuration (Graton and Fraser, 1935). A plot of intergranular volume against intergranular cement leads to the following results (Fig. 52):
It appears that up to 60% of the original porosity is apparently destroyed by mechanical compaction (chemical grain-contact dissolution is negligible in this sandstone). The higher porosity samples appear to be more compacted than the low porosity samples. The cementation causes a reduction of the original porosity of up to approximately 5%, assuming that there is no significant change in detrital grain size.

The porosity reduction in the Lochaline sandstone is always linked to the reduction in effective pore size as seen in Fig. 99.

What controls the 2D porosity?

The techniques employed have different resolutions. The SEM image is analysed by employing 128 transections, each 4.5 mm long, with a point space of 9 µm. The thin section point counting is based on four transections, 30 mm long, with a point spacing of 0.3 mm (equivalent to average grain diameter). The resulting areas of investigation are approximately the same (13.5 mm² for the image analysis and 54 mm² for point counting). Both 2D methods are limited by the choice of the "representative" area and by any anisotropy of pores. The later is thought to be negligible in this sandstone. The representative area was chosen by photography (Appendix 8.3) rather than obtained by a time consuming analysis of many randomly chosen images. It is not thought that a higher frequency of analysed images would have significantly affected and improved the results resulting from greater statistically stability. However, it is possible that higher magnifications of a single SEM-image could increase the average porosity.

Since the helium grain density shows values of approximately 2.64 g/cm³, which is very close to the density of a single quartz grain (2.647 g/cm³), one would expect that the total area (2D) porosity should equal the connected three-dimensional porosity. This only occurs for highly isotropic rocks and LSst is considerably isotropic. However, one must also take into consideration the discrepancy between the scale of investigation for the He porosity measurement (approximately 2000 mm²) and the 2D investigation (13.5 or 54 mm²).

The lower porosity values of SEM images are commonly referred to as micro-porosity due to clay minerals (Solymar, 1998), but this could not be confirmed for the clay-free Lochaline sandstone. The higher porosities of the point counting technique in comparison to the SEM image analysis technique are most likely caused by the "deeper" investigation into the 30 µm thick sample, in the former case. Nevertheless, the trend of overestimating 3D-porosity by point counting and underestimating by SEM image analysis (Fig.61) can not be explained sufficiently at this stage.
4.3 Permeability

4.3.1 Gas Permeability (Klinkenberg corrected)

The gas permeabilities of 21 samples were determined with N\textsubscript{2} and were corrected for the Klinkenberg effect.

The permeabilities of samples were comparable to each other within a specific series, but showed significant differences between series. The permeabilities range over several orders of magnitude, from 1 up to 2400 mD.

For a first overview, the values are presented in a bar chart in Fig. 64. All Klinkenberg plots, which show the raw data, are presented in Appendix 8.5, apart from three representative plots of the sample series in Fig. 63.

Samples of the O-series tended to be very permeable, having values between 466 mD and 2400 mD (Fig. 64). The sample with the lowest value (O1) include a large fracture marked by fine white grains. The correlation coefficients $R^2$ of the Klinkenberg plots are satisfactory; they range between 0.86 and 0.99. The permeability values over about 400 mD become less precise because the pressure difference from which the permeability is calculated falls.

Samples of the G-series showed intermediate permeability values between 53 mD and 878 mD, excluding the G20-sample, which has very low permeability (2.5 mD). These data were obtained from very good Klinkenberg regression trendlines with correlation coefficients between 0.99 and 0.98.

All samples of the H-series revealed a low permeability, varying between 1 mD and 68 mD. The values are derived from very good Klinkenberg trendlines ($R^2 = 0.98$).

The gas slippage factor increases exponential with permeability from the H-series to the O-
series. The trendline could be described by the following equation:

\[ \text{Gas slippage factor} = 1.23 \ e^{0.0018 \ K} \] where, \( K \) = permeability [mD] (Fig. )

It appears that the tubular burrows (in samples H4, G13, G15, G16, G17 nearly perpendicular to the gas flow) have no influence on the permeability.

![Graph of Klinkenberg permeability plots of three representative samples](image)

Fig. 63: Klinkenberg permeability plots of three representative samples
Fig. 64: Bar chart of Klinkenberg permeabilities of the Lochaline sandstone measured at confining pressures of 400 psi. The predicted permeability after Pittman (1992) and Katz & Thompson (1987) is also included. The raw data used to calculate the parameters of the Lochaline Sandstone samples of the LH- and LS-series were provided by Alzadjali (1997).

\[ y = 1.2315e^{0.0018x} \]

\[ R^2 = 0.9207 \]

4.3.1.1 Confining Pressure Dependence

The confining pressure was varied from 200 to 1800 psi in order to investigate its effect on the Klinkenberg air permeability. Due to technical limitations, the upper pressure limit was 1800 psi, with no backup pressure being applied. The dependence of permeability on confining pressure was examined on one representative core plug from each the high and low porosity formations (sample O8 and H5).

The low porosity sample (H5) showed a marked decrease in permeability after raising the confining pressure from 200 psi to 400 psi (Fig. 65). The decrease of about 0.5 mD probably reflects sealing between the core plug and core holder chamber. As expected, a further increase of confining pressure insignificantly affects insignificantly the microstructure.
However, a tendency of decreasing permeability with an increase in confining pressure exists due to rock deformation processes and consequent reduction in effective pore size (and/or change in tortuosity). The slight increase in permeability at 1400 psi could be due to grain rearrangements. The difference between the initial permeability and the final permeability at the end of the relaxation process indicates inelastic sandstone deformation processes.

The highly porous sample O8 reveals a marked decrease in permeability from raising the confining pressure to 600 psi. The subsequent, significant increase in permeability, after increasing the pressures up to 1000 psi, is probably caused by the formation of micro cracks and/or fractures. A further increase in the confining pressure up to 1200 psi resulted in formation of fractures and finally (15 minutes after adjustment) in the collapse of the core plug.

A photograph of the sample, prepared perpendicular to its axis after the collapse, is shown in Fig. 66. The sample shows fractured grains and grains containing micro-cracks. Because examinations of other samples, which are not affected by pressure, revealed similar textures of crushed grains, the observation cannot solely be attributed to pressure effects. However, results of sandstone deformation studies suggested the onset of grain crushing at pressures as low as 980 psi (Zoback and Byerlee, 1976).
Fig. 65: Plot of Klinkenberg permeability versus confining pressure Pc for a cemented (H5) and uncemented (O8) sample.

Fig. 66: Micrograph of sample O6 after applying a confining pressure of 1200 psi. Fractures in grains caused by the compressive strain are arranged in two directions.
4.3.2 Probe Permeability

Probe permeametry was carried out on a wall within the Lochaline sand mine using a 2 man probe permeameter. A vertical mine wall was chosen for the measurements which included a minor fault. The dimensions of wall measured were 3.6 m long and 1.5 m high. Measurements were taken on a grid with a approximate spacing of 0.5 m at points which had been prepared for probe permeametry by a previous study (Lewis et al., 1990). Preparation of the measurement points included shallow drilling to remove the disturbed top layer of rock, followed by milling the bottom of the hole flat to accept the testing head of the probe permeameter. Figs. 70 & 71 show the results and location of the probe permeametry. It should be noted that the top right hand corner of the area could not be accessed for measurements due to safety considerations.

The measured relative permeabilities range from approximately 1000 mD up to 6000 mD (Fig. 70). The formation shows significantly different permeabilities on either side of the fault, which is located between 200 and 250 cm on the x-axis (indicated by the lack of sampling points). The unusually high permeability of the formation towards the right hand side of the sampled area appears to be caused by the fault. Almost every vertical sampling section shows an increase in permeability, which could reflect a coarsening-upward sequence. Whilst it is known that permeability variation could relate to grain size, no such sedimentological explanation was found at the examined wall. The only detected feature was horizontal iron-staining (Fig. 71), which has no significant influence on the permeability.

A plot of Klinkenberg permeability versus probe permeameter permeability obtained from measurements on prepared Lochaline Sandstone core plugs reveals comparable values for the two different methods (Fig. 69). The values vary within one order of magnitude, which is typical of the accuracy obtained from probe permeametry. A trendline drawn through the points shows a satisfying correlation coefficient of 0.61.
Fig. 69: Plot of permeability data measured by mini permeameter versus permeability derived from routine core analysis.

\[
y = 0.7449x + 1 \\
R^2 = 0.6128
\]

Fig. 70: The upper layer shows the sampling points and the permeability zonations. The lower layer presents the interpolated permeability variation in a relief plot. The x-axis represents the horizontal orientation, whereas the y-axis represents the vertical length.
Fig. 71: Photograph of the left part of the sampling area. Horizontal iron staining is visible.

4.3.3 Brine Permeability

The brine permeabilities of samples G17, G19, H3, O3 and O4 were measured in a fluid flow rig at 500 psi confining pressure. The bulk of the samples show slightly lower brine permeabilities in comparison to the Klinkenberg air permeabilities. This difference is expected since the confining pressure was 100 psi higher. Only the very low permeable sample H3 reveals a brine permeability, which is significantly higher than the Klinkenberg gas permeability. The error bars indicate the maximal and the minimal measured value at different flow rates.

Fig. 72: Plot of brine permeability versus Klinkenberg permeability.
4. 3. 4 Predicted Permeability

The correlation between calculated Klinkenberg permeability and parameters obtained from mercury injection, helium porosimetry and electrical measurements are presented in two ways.

Firstly, a comparison is made between fictive trendlines in a poroperm cross plot created by different models under employment of fixed parameters. The measured poroperm data were presented in those plots as well. The purpose of this exercise is to access whether data and model trends are in general agreement or not.

In the second step, predicted permeability values based on defined parameters were plotted against measured permeabilities. Some of the models introduced in Chapter 1.3 are employed to predict permeability; namely, the pore size models of Pittman, Swanson, Kozeny-Carman, Katz & Thompson and the grain size models of Berg, Van Baaren and Revil et al..

Pore Size Models

A comparison of the Katz & Thompson model with the apex-model of Pittman reveals similar poroperm trends of constant pore sizes. At high porosities (>10 %), it is observed only an insignificant change in permeability with decrease in porosity. This trend is in agreement with the data trend. Both models and the data show massive decrease of permeability at lower porosities. However, the poroperm trend of the data measured shows a more advanced decrease of permeability with porosity at porosities below 10 percent, which can not be described by a single porosity power law. The data trend crosses the model trendlines of constant effective pore radii (fig.73). This could suggest a massive decrease in the effective radii, explained by the increasing degree of cementation. One should note that neither the data nor the models fit to a straight exponential line in a poroperm plot, which is commonly used in the oil industry to characterise reservoirs.

Plots of predicted permeability versus measured permeability reveal that models tends to underestimate permeability except for the lower permeable samples where they predict too high permeability values (fig.75). Swanson’s and Pittman’s apex model is based on parameters gained from the same location, the second inflection point of the capillary pressure curve or respectively radii curve. However, Swanson’s method significantly overestimates the measured permeability, Pittman’s apex method yields to predicted values, which fall within plus/minus a factor of ten of the measured permeability. Both models appear to overestimate permeability in the low permeable samples. One should note that the equations of both models are based on multiple regression analysis of uncorrected air permeabilities.

Models including a fixed additional electric parameter show better correlations with measured permeabilities. The predicted permeability after Katz and Thompson was calculated in two
ways. The first data set consists of values derived from m - values of each single sample. For the second data set a single m-value was chosen which represents the entire range of data (m=1.8) and the average grain diameter from mercury injection was incorporated (D = 590 µm). This second data set revealed a much closer correlation to the measured permeability. Again, it is observed an underestimation of permeability over almost the entire data set.

The permeability was also calculated in two ways for the Kozeny-Carmen model. First it was employed the pore surface area derived from mercury injection normalised by the bulk sample volume. Nevertheless, this specific surface can also be represented by the reciprocal value of the "hydraulic" radius. The Kozeny-Carmen permeability was calculated a second time by incorporation of the hydraulic radius. The "hydraulic" radius is thought to be equivalent to Katz and Thompson effective radius at the entry pressure of the capillary curve. The Kozeny-Carman model overestimates permeability over almost entire data set but correlates very good with the measured permeability. Permeabilities predicted under incorporation of the specific surface are closer to the measured permeability than those derived from the effective radius.

Grain Size Models

All three models show similar poroperm trends of constant grain diameters. Those permeability models are very sensitive to changes in porosity; they relate permeability to the power of five of porosity.

The trendlines fit to the data points of the low porous Lochaline Sandstone samples very well, which are influenced massively by cement evolution. The uncemented samples of the O-series could be modelled by a smaller grain size. However, it is not thought that the cementation affects the grain size significantly (see Chapter 4.1.3.). The reduced permeability of the O-series could also be caused by slightly poorer sorting (fig.42), which could result in blocking of pore throats by smaller grains or by higher degree of compaction (fig.52).

In all models, it is an input grain diameter of at least 1200 micron necessary to obtain poroperm trends, which fit the data. This figure is significantly higher than the average grain diameter of the Lochaline Sandstone of 238 micron, as determined by Humphries (1961). Therefore, the predicted permeability resulting from measured grain diameters would be much too low in comparison to the real data - as indicated by the poroperm trendline of constant grain size of 250 µm (fig.74). The results suggests that there are more permeable zones within the sandstone samples than assumed by the restricted consideration of grain packing, e.g. due to oversized pores or fractures, which could increase the intergranular pathways.

Predicted permeability derived from the RGPZ model under incorporation of the average grain size from mercury injection and a constant m-value of 1.8 underestimate permeability in the G and H-series within one order of magnitude (fig.75). However, it predicts permeability for the samples of the O-series very well. Prediction of permeability, carried out by
incorporation of sample specific m-values and grain sizes from mercury injection, result in a weak correlation with measured permeability (fig. 75).
Fig. 74: Poroperm trendline of constant grain size in microns calculated from the three grain size models. It was taken a representative m-value of 1.8 for the Lochaline Sandstone to calculate the RGPZ and the Van Baaren trendlines. The points are measured data of Lochaline Sandstone core plugs.
Fig. 75: Plot of predicted permeability (y-axis) versus measured permeability (x-axis).
4.3.5 Controls

The controlling factors in permeability of the Lochaline Sandstone are figured out by crossplots of permeability versus helium porosity, specific surface from mercury injection, electrical tortuosity and characteristic pore size parameters. Permeability relates best with porosity as indicated by the very good correlation coefficient of 0.94. The entire poroperm trend can be satisfying modelled by a porosity power law of 3.8. Sample O1 reveals too low permeability to fit to the trendlines due to healed fractures (extreme fine grain size) across the core plug.

However, in order to improve the fit of trendline with the sample points, the samples are divided into a high and a low porous group. The highly porous O-series shows too high porosities or too low permeabilities to fit the trendline of the low porous samples. The permeability could be reduced due to mechanical compaction with grain fracturing (could block pore throats), as observed in the O-series. It is not thought that the insignificant increase in grainsize during cementation will influence the poroperm trend. The poroperm trend, which represents only the low porous samples (<12 %) shows a better correlation coefficient (0.97) than the one which represents the entire range of samples. The permeability of the low porous samples reacts very sensitive to any change in porosity as indicated by a power law of 5.5.

The less accurate mercury porosity shows a weaker poroperm trend (correlation coefficient = 0.78). However, there exists an improvement of the poroperm trend under exclusion of the ineffective fractal porosity as seen in figure 77.

The high permeable samples (>100 mD) are also related to the effective pore throat radii of Katz and Thompson (1987) as well as to the pore radii of Pittman (1992). It is observed a decrease in pore size with decreasing permeability, which follows a power law of 3.2 - 3.5. The pathways in the low permeable samples appear to be not affected by effective pore radii, since they show no significant change below 100 mD.

The influence of the complexity of the flow path can be examined by cross plots of permeability versus the pore body / pore throat ratio and versus the electrical tortuosity. The ratio of pore body (derived from SEM feature analysis) to pore throat (related to apex) shows a very good correlation with permeability by a power law of -2.3. An increase in the relative difference of the average dimensions of pore body to pore throat relates to a decrease in permeability, in particular in the low porous samples (<100 mD). An increase in complexity of the pathway is also indicated by an increase in tortuosity. The tortuosity shows an expected trend of higher values with decreasing permeability.

The specific surface, the total wetted surface, is primarily controlled by the grain size (small grains – large specific surface). One would expect a decreasing in specific surface with increasing permeability. However, the trend in figure 78 reveals a reverse relationship. The small variation in specific surface is suggested to be only a secondary factor in controlling permeability in this sandstone. But the higher specific surfaces of the high permeable samples
could affect the poroperm trend, as seen in the reduced permeability values of the high porous samples in figure 76 which results in a different poroperm trend.
One should bear in mind that a single process massively affects all the petrophysical parameters of the low porous samples: cementation.

Fig.76: Poroperm trend of the Lochaline Sandstone.

Fig.77: Left chart: Plot of mercury porosity versus permeability.
Right chart: Plot of Euclidean mercury porosity versus permeability.
Fig. 78: Plot of permeability versus petrophysical parameters.
4. 4 Electrical Properties

All electrical parameters were calculated from resistivity measurements. The calculation of the electrical tortuosity and the cementation factor incorporate also the helium porosities.

4. 4. 1 Formation Factor

The formation factor was obtained by the ratio of rock resistivity to fluid resistivity at a frequency of 1kHz. Measurements were carried out on 16 samples. The values of the formation factor ranging from 7 up to 245. The formation factor increases from the O-series (7-15) over the G-series (27-124) to the H-series (75-245). The data plotted against porosity reveal a power law behaviour as represented by Archie’s equation which relates porosity to formation factor. The dashed line in the plot represents Archie’s relationship of formation factor to porosity by a power of two. One of the first attempts to duplicate the cementation process by injection of silicate gel and subsequent heating was carried out by Wyllie and Gregory (1953). They concluded to add a constant to Archie’s equation, which in the case of the Lochaline sandstone equals 0.62, and which should be controlled by the sample's pore system before cementation. Their equation was chosen in order to obtain a better fit to the measured data set. The exponent of Wyllie and Gregory’s equation could be derived from the gradient of a trendline drawn through the data set. It is given in the case of the Lochaline sandstone formation by 1.88, which fits the entire range of data very well. Nevertheless one should bear in mind that under in situ overburden conditions a massive increase in the exponent of Wyllie and Gregory’s equation as well as of the exponent in Archie’s equation will occur (Fatt, 1957).

There can not be seen any evidence of pore throat closing during cementation, which would result in an upward trend in the low porosity tail of the data in the log-log plot of formation factor against porosity in figure 79 (after the grain consolidation model of Roberts and Schwarz, 1985).

4. 4. 1. 1 Salinity Dependence

For sample H5 and O8 was measured the dependence of the conductivity as a function of brine salinity at 1 kHz and confining pressures of 500 psi. The salinity was subsequent decreased from a two mol NaCl solution to distilled water within the fluid flow rig. The pump rate was adjusted to two cubic centimetres per minute and manual controlled.

The change in impedance was recorded as an average of five readings every 125 ms and is presented in figure 81. The x - axis represents the time after changing of the fluid pumped. Both samples reveal no significant difference in the time the "tracer" needs to penetrate the sample, both of them show an increase in resistivity after approximately 8 minutes. Moreover the fluid pumped through the high porous samples O8 appears to require slightly more time to the first inflection point of the break trough curve. As well, the fluid requires more time to completely saturate the plug. These findings are in contradiction to the much higher electrical
tortuosity of the low porosity samples (Chapter 4.4.3), if one defines tortuosity by measuring
the time required by ions to pass through the pores. Hence, the pore volume appears to affect
more the saturation process than the complexity of the pore system. The apparent steady
increase in resistance with time in the low porous sample H5 could be explained by the high
streaming potential.

Both samples reveal problems at the determination of the resistance at the very low salinities
of 0.00001 mol NaCl and at distilled water. The measurement at distilled water is thought to
be significant affected by the measurement apparatus and so excluded from the conductivity
plot.

Plotted the samples conductivity as function of its fluid salinity after 2000 seconds both
samples show an almost straight line relationship over the entire range of salinities (fig.82).
This is expected since the sandstone contains no clay minerals and surface conduction
processes are very limited. The surface conductivity is calculated by the subtraction of the
ratio of fluid conductance to formation factor from the total conductivity. However, the
formation factor was determined in the high electrolyte conductivity regime (1mol NaCl) to
avoid any surface conducting effects. The resulting surface conduction is approximately
0.0001 ohm.m for both samples which agrees with the experimental values obtained from the
clay free Fontainebleau sandstone (Ruffet et. al., 1991) and with theoretical surface
conduction models (Revil and Glover, 1997). One should note that the data points at the
lowest fluid saturation could contain a large error since they were measured with a sample
saturated with a 0.00001 mol NaCl brine.

4. 4. 2  Ceme ntation Factor
The cementation factor m was determined for each single core plug. The data reveal slightly
decreasing m-values with increasing porosity. The values are ranging from 1.8 to 1.45. The
lower limit from the uncemented formation is in agreement with m-values of 1.5 from
experiments of packs of sperical grains of many different sizes carried out by Sen et al.
(1981). The higher m-values are probably the result of small changes in the pore space
distribution. The continuously higher m-values at lower porosities suggests that the
conductivity reacts slightly more sensitive to any change in porosity - small changes in
porosity leads to huge changes in conduction. Since the formation factor is related to porosity
with a power law, in which the cementation factor acts as exponent, one could suggest that
with decreasing porosity some large pores remain stable while throats gradually shrink and
become eliminated. The nodes are pocket-like (see Chapter 4.6.1.2.) and will contribute much
to the porosity but little to transport (permeability, conductivity). They add to the skewness of
the pore space distribution and will therefore increase the m-value.
4.4.3 Tortuosity
The electrical tortuosity is ranging from 1.6 to 10.5. A plot of the values against porosities reveal an increasing trend of the tortuosity with decrease in porosity, meaning that the flow path created during cementation becomes more complicated. The trend follows a power law of 0.88 and appears to approach a threshold at around two-three percent porosity. It is suggested that the percolating flow stop at this "cut-off" porosity value in the Lochaline Sandstone. It has also been proven theoretically by Van der Marck (1996) that a threshold exists at 3% porosity for electrical conduction for various packing of grains with different sizes. For the genetic similar Fontainebleau sandstone was recently proposed a percolation threshold for permeability at 2.5 percent porosity (Mavko and Nur, 1997). The subtraction of the "ineffective" part of the porosity (below the threshold) from the total porosity is suggested to improve results obtained from well established equations like Kozeny-Carman’s equation or Archie’s equation (Mavko and Nur, 1997; Van der Marck and Hausenblas, 1997).

4.4.4 Controls
The question of the controlling factors in electrical conductivity is of particular interest since quantitative parameters like electrical tortuosity or "effective" pore radii are tried to be transferred to fluid flow transport processes in order to predict e.g. permeability. As we have seen, all electrical properties are correlating very well with the porosity. The inverse formation factor as presented in figure 79 in semi - log plot against porosity can be seen as a normalised parameter to describe conductivity. The plot reveals clearly a simple correlation between conduction and porosity which can be described in a single power law equation (R-squared = 0.96). One should note that the poroperm relation was more complicated. This confirms that the shape and size of grains in non shaly rock insignificantly influences electrical properties as already proven empirically (Archie, 1952) and theoretically (Cohen, 1981) but considerable influences permeability. A plot of normalised electrical conductivity (1 / FF) versus permeability shows a good correlation and relates conductivity to permeability by a power law (fig.80). This is expected since the electric conductivity scales to an effective pore radii power of two whereas the permeability scales to an effective radii with a power of four (David, 1990). In the case of the Lochaline Sandstone, the conductivity scales to the effective apex pore radii power of 2.5 (fig.81) and the permeability to an effective pore radii power of 3.5 (fig.78).

The observed increase in tortuosity could not only be affected by decreasing porosity but also by higher complexity and/or longer path way (decrease in effective porosity). As seen in figure 81, the time of the first breakthrough of a different saline brine appears to be not significant different and so the path length of the low and the high permeable sample. The SEM - image photographs confirm this assumption. They reveal little evidence of complete pore throat closure during cementation (appendix 8.3). The degree of constructivity of the path caused by pore constrictions was approximated by the ratio of pore body to pore throat of each
single sample. The "effective" Katz and Thompson pore radii were taken as the parameters for the pore throats and the median pore radii of the area weighted SEM pore radii sum curve were employed as the parameter which represents the pore bodies. Figure 79 shows the plot of the constructivity against tortuosity. It is observed only a weak relationship, as expected from the inaccurate determination of the ratio as well as from the assumption that in general the tortuosity depends on both, the constructivity and the length of the path. Nevertheless, it is suggested that the tortuosity of the cemented samples of the sandstone be mainly controlled by the constructivity of the pore system and less by differences in the path length. One should also note that results from numerical simulations suggest a hydraulic tortuosity which is 1.5 times higher than the tortuosity for electrical current flow for sands with highly skewed pore size distributions (David, 1993). But he suggested also that the error introduced by using the lower electrical tortuosities, in models of permeability prediction, may be compensated by employing simultaneously the lower hydraulic radius (equivalent to the pore entry radius), e.g. in the well known Kozeny-Carmen equation, in place of an appropriate larger length scale for fluid flow.

The **m-value**, also called cementation factor shows a consistently increase with decreasing porosity. From pore space models and observation in synthetic porous media, it is known that the m-value is highest when the shrinking produce a wide range of pore sizes and the m-value is low in the case of pores of the same size (Wong et al., 1984). From measurements of carbonate cores it is also known that the m-value is affected by the presence of unconnected porosity - the higher the proportion unconnected porosity to total porosity the higher the m-value (Lucia, 1983). Because the cementation is the main factor which influenced the porosity in this rock and the relation of porosity to conductivity, the following two scenarios are possible: cementation increases the skewness of the pore size distribution or/and it increases the ratio of non-connected to total porosity.
Results

Fig. 79: Crossplots reveal the very good correlation between electrical parameters and

\[ y = 0.6244x^{-0.8838} \]

\[ R^2 = 0.9674 \]

\[ y = 1.4042x^{1.471} \]

\[ R^2 = 0.8878 \]
Fig. 80: Crossplots of electrical conductivity (1/FF) versus hydraulic conductivity, porosity and characteristic radii. Particularly the low porous samples show a disproportional decrease of hydraulic conductivity in comparison to the electrical conductivity. The porosity-electrical conductivity trend follows a single power law.
Fig. 81:
Fig. 82: Crossplots of rock conductivity versus fluid conductivity. The surface conductance is in both samples very low (~0.0001 ohm.m) as expected from a clay-free sandstone.

4.5 Complex Dielectric Properties

The complex resistivity measurements were carried out on sandstone samples H1 - H4, G14, G20, G21 and O1 - O7. The complex electrical behaviour of a rock when submitted to an alternating electrical field at different frequencies results from both its conductive and dielectric response. The former is related to the transport of free charges and the latter is associated with polarisation at interfaces. This interface polarisation in the frequency range from 20 kHz to 500 kHz could be related to pore-scale parameters like pore geometry and specific surface, which in turn is related to permeability through the well-known Kozeny-Carmen relationship.

4.5.1 Argand Plots

The frequency dependence of the imaginary and real part of the complex resistivity is more pronounced at the low porous samples. The Argand plots seem to be composed of 2 semicircles. Polarisation processes between electrode and sample have larger time constants than
Those of the polarisation processes in the electrolyte or in the bulk of the samples under test (Knight and Nur, 1987). Therefore, it is thought that the lower frequency dispersion in the Argand plot corresponds to the electrode, whereas the depressed semicircular arc in the high frequency regime (left intimated circle) can be attributed to material properties and exhibits a Cole-Cole resistivity response.

In the higher porous samples of the G- and O-series can be observed almost no frequency dependence (fig.83). This could be explained by means of the relative high fluid concentration and the high conductive pore structure at which the dielectric response is negligible in comparison to the transport of free charges through the fluid.

Because it is difficult to determine the Cole - Cole constant from the not very prominent semicircular in the Argand plot, the constant is determined in a plot of frequency versus permittivity, from the slope of the real part of the relative dielectric permittivity at high frequencies (next Chapter).

4.5.2 Frequency Plots

Because Argand plots are not suitable to describe the frequency behaviour of the very high porous samples of the G- and O-series, the real and the imaginary part of the complex impedance and permittivity are plotted separately against the frequency.

The critical frequency at which electrode polarisation becomes insignificant compared to bulk sample response is marked by increasingly higher values of the imaginary part of the resistance at high frequencies (fig.84a/b). In few highly porous samples of the O-series (O1-3 and O6-7) becomes the imaginary part of the resistance “negative” at high frequencies. In most of the samples is the critical frequency, which separates the two semi-circles in the Argand plot, at approximately 20 kHz. Less polarisation effects are observed in the samples saturated in the fluid flow rig in the low frequency range because of the platinum net renew between sample and electrode previously to these measurements.

Samples, which were saturated in the fluid flow rig (H2, H4), reveal slightly lower resistivities than the samples saturated under vacuum (H2v, H4v). This could be explained by a higher degree of saturation of these very tight samples, which is resulting from high pressures used in the fluid flow rig. It is not thought that there is a conducting fluid film the high confining pressure applied (400 psi) between sleeve and core plug.

For practical reasons, permittivity plots are used to calculate the Cole-Cole exponent q. In the low permeable samples (<100mD), the results agree with a Cole-Cole behavior in the high frequency range (>20kHz): $\kappa'$ varies as $\omega^{-q}$ (power law relationship). The q-values are ranging from 0.047-0.354. The unusual high values compared to the results of Glover et al. (1994) reflect the higher fluid concentration. In the low frequency region (<1kHz), the response is characterised by a very strong frequency dependance of $\kappa'$ (fig.84c). The electrode polarization phenomena are very important in this frequency range and add to the bulk sample effects, so it enables not to characterize the low-frequency dispersion.
4.5.3 Controls

As stated previously, the frequency dependence of the resistivity was attributed to interfacial effects by many researchers. Knight and Nur (1987) presented a correlation between the power law exponent of 8 samples taken from the log-log plot of relative permittivity versus frequency, and the specific surface area in sandstone samples. Kuhlenkampff (1994) observed power law exponents taken from log-log plots of complex conductivity against frequency and correlate it with specific surface values (BET). Denicol and Jing (1996) correlate power law exponent taken from a semi-log plot of the impedance versus frequency with Klinkenberg air permeability.

In this study, the Cole-Cole exponent \( q \) is taken from the real part of the complex permittivity at high frequencies (\( > 20 \text{ kHz} \)). In the case of the Lochaline Sandstone, the \( q \)-value correlates very well with the permeability (fig.85), which results in a correlation coefficient of 0.94. The plot reveals the following power law relationship between the two parameters:

\[
K = 1813.6 \, q^{2.1832}
\]

where

- \( q \) = Cole-Cole exponent
- \( K \) = Klinkenberg Permeability [mD]

This result is in agreement with the observations of Denicol and Jing (1996) which have obtained also a very good correlation coefficient of 0.91 between the two parameters. One should note that the calculated Cole-Cole exponent of the low permeable samples (\(<100\text{mD}\)) reveals realistic \( q \)-values below the theoretically upper limit of 0.5.

The Cole-Cole exponent showed a weak correlation with the specific surface normalised by the bulk volume (correlation coefficient = 0.4). This observation is the contrary of results of Ruffet et al. (1991) who observed a clear correlation between \( q \)-value and the specific surface of the samples, but it is in agreement with a weak correlations coefficient (0.4) obtained by Kuhlenkampff (1994, p.121). The weak correlation is unexpected since the specific surface serves as a theoretical link between permeability and \( q \)-value. It could be explained by the relative inaccurate determination of the specific surface by the mercury method. It could also be explained by the assumption that the large changes in grain and pore geometry affects more the degree of polarisation than the small differences in specific surface between the samples. However, both parameters, the specific surface as well as the \( q \)-exponent, are positively correlated to permeability (fig.85).
Fig. 83:
Fig. 84a:
Fig. 84b:
Fig. 85: The specific surface and the q-value show a positive correlation with permeability.

\[ y = 1813.6x^{2.1832} \]
\[ R^2 = 0.9417 \]

\[ y = 8971.6x^{4.4922} \]
\[ R^2 = 0.9632 \]
4. 6 Pore Geometry
Three methods were applied to investigate the change in pore geometry resulting from
diagenesis: direct observations of pore casts and broken surfaces by the SEM of sample O6,
G16, H1 and H3 and indirect measurement of the electrical conduction and the capillary
pressure behaviour. Quantitative statements were obtained from complete pore fraction size
distribution (from mercury injection and SEM feature analysis), fractal dimension of the grain
surface (from SEM, mercury injection data and complex resistivity measurements) and
characteristic pore radii (from SEM and mercury injection).

4. 6. 1 Qualitative Characterisation
4. 6. 1. 1 Capillary Pressure Curves (appendix 8.2)
Despite the pore system of a sandstone is complex, the shape of the capillary pressure curve
provides significant insight into the nature of the samples pore geometry. The pressure curves
have three distinct characteristics in the case of the Lochaline Sandstone: the first inflection
point - called entry pressure, the second inflection point - called threshold apex pressure and
the "plateau" shape part of the curve between the two points (fig86).
All samples show extremely low entry pressures ranging from approximately 5 psi (O-series)
up to 15 psi (G- and H-series). The threshold pressure saturation shows a large variety
between the series. The O-series reveals extremely unusual high threshold-apex saturations
ranging between 75 and 60%. This could be explained by a very homogeneous and well
connected pore structure - only a small increase in pressure was necessary to saturate most of
the pore space. Decreasing threshold saturation from 80 to 45% with decreasing porosity is
observed in samples of the G-series. The H-series shows the lowest mercury saturation at the
threshold apex pressure ranging from 35 to 60%. The broad variety in the H-series, which
appears to be independent of porosity, results from diverse cementation processes. In some
samples, marked by the still high threshold saturation, it is expected that the complexity of the
pore structure (the ratio pore body/pore throat) was minor affected by cementation. The
samples with the lower saturations at the threshold pressure are thought to contain large pore
bodies, which are not connected to the first interconnected path through the sample. Only few
of those pores can massivly reduce the degree of saturation in the low porous samples. One
should note that the average apex-saturation is around 35%. The mean mercury saturation for
the threshold pressure of Pittman’s (1992) data set was 36 %, and Winland found best
correlations with radii related to 35% saturation (Kolodzie, 1980). Roberts and Lin (1996)
found in conductivity measurements that interconnected fluid in Tuff samples of 9 - 28%
porosity exists also at 35% saturation.
The “plateau” of the capillary pressure curves represent an important part of the pore network,
which dominates mercury saturation. It is developed very prominent and nearly horizontal in
all samples of the O-series. This feature indicated that the pores were relatively uniform in
size as expected (corresponding to spherical grains). Additionally, it could reflect a
homogeneous pore throat distribution. The relatively low plateau levels corresponded with small entry pressures. The plots of pore radii versus mercury saturation of the O-series showed that almost no pore volume was isolated by pore apertures of less than 1 micron radius, which would correspond to an excellent reservoir rock.

The plateau of the curve in a pressure versus saturation plot from the samples of the G- and H-series was positioned at slightly higher pressures and less flat (appendix 8.2) than in the O-series, indicating a slightly more heterogeneous pore throat distribution. The entry pressures, which are related to a small fraction of the pore volume containing the largest pores, are slightly higher than in the samples of the O-series.

Despite the drainage curve was not possible to recover, the degree of fluid recovery is an important feature. In the original raw data prints can be seen the hysteresis effect (Bächle, 1997), which occurred almost in all samples due to the high capillary force in narrow pore throats which did not allow the drainage of larger pores. This fact suggests that the ratio (in the smaller pores size range) between size of pores and pore throats seemed to be high, maybe caused by oversized pores due to solution. Only the samples G14 and O2 showed complete drainage of their pores (low pore body / pore throat size ratio), while sample H4 revealed the least effective drainage.

To investigate further how oversized pore bodies in the low porous samples of the H-series affect the capillary pressure curve, a secondary injection cycle after drainage was carried out in one sample of the H-series and one of O-series (fig.87). The co-ordination number, the average number of throats, which are connected with single pores, and the roughness of pore surfaces is thought to changes insignificantly with cementation and hence will have a negligible influence on the trapping behaviour. The saturation at the first possible pressure reading point is seen to be roughly equivalent to the irreversible mercury saturation. The low porous samples reveal a lower degree of mercury recovery (~60%) compared with the O-series (~80%). However, the recovery efficiency of both samples is very high. The apex-threshold saturation in a second injection circle increases in the low porous sample H5 and decreases in the high porous sample O8. In the low porous sample decreases also the apex-threshold pore radius. The effective radii in sample H5 remain constant. Below 80% mercury saturation, there is less pressure necessary to saturate the pores (the large pores connected by small pore throats are now included), but there is more pressure necessary to fully saturate the sample. This will result in a more realistic pore size distribution, with a higher percentage of larger pores and smaller pores, than measured during routine mercury injection. Liu and Winslow (1995) revealed that the pore entry diameter of such reversible sub-distribution, derived from a second intrusion cycle, were more closely related to permeability.

One should note that in the highly porous sample which has a much better connected pore system, the secondary intrusion resulted in a decrease of pore sizes over the entire pore size distribution range. Irreversible compaction effects during the first injection/drainage cycle could explain this observation.
Fig. 86: Capillary pressure curves of four samples of the Lochaline Sandstone. One should note the extreme high mercury saturations at the second inflection points (related to apex pressure).

Fig. 87: Capillary pressure curves of a routine (first injection) and repeated (second injection) core analysis circle of a low and a high porous sample of the Lochaline Sandstone. The low porous sample (H5) shows a marked higher apex mercury saturation (second inflection point) after repeated mercury injection.
4.6.1.2 SEM Images
Electron microscopy images are obtained from polished thin section surfaces of 14 samples, whereas three-dimensional images of the pore structure and of the "negative" pore casts are taken only from four representative samples (O6, H3, H1 and G16). Since mainly quartz overgrowth determined the pore geometry of the G- and H-series, the pore throats geometry is dominated by flat slot-like pores (fig.91) and, in less well connected areas where grains may had welded together, by narrow tubes (fig.92). The pores were smaller and polygonal (fig.93) because of the euhedral quartz development, in comparison to the smooth and well interconnected pores of the O-series (fig.88). It appears that even between highly cemented grains still remains open pore space as seen in the two dimensional SEM-images (appendix 8.3), marked by the black lining which represents pore space. Maximal thicknesses of this pore space of approximately 3-5 µm are measured from high magnification images of grain boundary.

The two-dimensional images show not only a reduction in dominant pore size with cementation but also a reduction in the absolute amount of large pores within a defined area. One should note that oversized vuggs, which were observed in low as well as in high porous samples, were tried to avoid in the examined area because it was thought that the resulting porosity would not have been representative for the bulk sample. Despite the oversized pores were not represented by the photographs, the observed existing oversized pores are much larger than the largest pores detected by the mercury injection method. Solution processes and authigenic illit formation had negligible influenced the pore sizes at micro-scale. However, few rare micro pores were detected by SEM observations (fig. 36, 92 and 94), but they appear not to be restricted to cemented samples. The significantly higher percentage of "micro porosity" on the total porosity in the low porous samples as detected by mercury injection could be caused by macro pores connected by very small pore throats.

4.6.2 Quantitative Characterisation
4.6.2.1 Apparent Pore Size Distribution
In order to ensure the cover of the entire range of pore sizes, two different techniques were employed. The SEM feature analysis provides data on the frequency and size of pores. Compared to the mercury method, which results in pore size estimations of a narrow, small pore throat size range, the SEM includes information of pores of a broader pore size spectrum. The first part will cover the "real" distribution of pores, weighted by their occurrence on the SEM image. The second part will include pore sizes weighted by the pore volume from mercury injection respectively weighted by the pore area in the case of the SEM image analysis. One should note that the capillary pressure required to invade a pore is governed by its pore throat radius while the volume intruded into the pore is controlled by the pore body radius. Since no firm correlation exists between pore throat and pore body radii in the 3D
case, the interpretation of the capillary pressure curves in terms of pore throat distribution is a weak one.

SEM - pore size distribution weighted by the number of features analysed
The pore size distribution is positively skewed towards the small pore size for all analysed samples of the O-series and the highly cemented sample H3. More than 25% of their pores are smaller than 6 µm in diameter, whereas only 10 - 20% of the pores of the other samples are smaller than 6 µm. A less pronounced skew of the pore distribution reveals sample H2, H5 and G13. The other samples of the G-series and sample H1 and H4 reveal no predominance of small pores histograms of pore fraction size based on the logarithm to a base of two (fig.95). The area sum curve, which is also plotted into the charts, reveals to which degree each pore size fraction contributes to the total SEM-porosity.

In general, homogenisation and continuous decrease of large pores occurs with cementation from the O-series to the G- and H-series, as observed by the left shift in the large pore size range and by the more linear sum curve in a semi-logarithmic plot of the frequency of the number of analysed pores versus pore diameter (appendix 8.3). No statement is possible to the evolution of the very small pores (< 4.3 µm) at the applied scale of image-magnification. One should also note that the occurrence of the absolute number of pores is significant decreasing with decreasing porosity from over 900 to only 200 analysed features.

Measurements of the maximal diameter of the pores monitor the evolution of the pore shape during cementation. Plots of the sum curve of mean and maximal diameter reveals that all pores detected are larger than 8 µm in maximal diameter (appendix 8.3). With the exception of sample H4 the number of analysed features in the smallest bin class 8 - 10 µm equals approximately the number of features analysed for the smallest detected mean diameter pore class. Either the smallest pores consist of the same mean-max pore diameter ratio which leads to the conclusion that the very small pores are very elongated or the smallest features detected are only artefacts. However, one should note that a filter, used to avoid artefacts, has already eliminated approximately half of the total amount of features counted at each sample.

A significant change can be observed neither in the relation of the maximal to the mean sum curves of the different samples nor in the elongation of pore radii within single samples. The large pores in the O-series appear to be more spherical. Only sample H3 reveals significant more elongated pores over the entire range. This leads to the assumption that cementation will not significantly influence the pore shapes, which are in all samples more elongated at smaller pores.

Hg- respectively SEM- pore size distribution weighted by pore volume (respectively area)
The results are presented in three ways, by complete double logarithmic SEM-Hg pore fraction distribution histograms weighted by volume or respectively area, by plotting the
mercury pore throat radii frequency curve and by plotting the sum curve of the SEM pore diameter curve.

The double logarithmic pore fraction plots are used for a rough overview to detect patterns of the evolution of pore distribution during cementation as well as for the determination of the entire occurring pore size range. The pores are ranging from 0.01 up to 1400 µm in diameter. The charts reveal that small pores disappear during cementation. Both techniques show a skewed, unimodal distribution towards larger pores, most advanced in the uncemented samples of the O-series (fig.96). The observed dominant pores with the SEM-tool are almost one order of magnitude larger. The overall shape of the samples pore distribution histograms of the mercury method, corrected by the “shift” in predominant pore size, pass into the SEM pore distribution histograms very well.

All the large pores, which are obviously not represented by the mercury pore distribution, are included in smaller pore throat fractions. The weakness of estimating pore throat size distribution by means of mercury injection under employment of the capillary tube model lies in the assumption that all pores are equally accessible to the invading phase. Particular in the highly cemented H -series, it is much likely that the pore volume of large inaccessible pores (as seen e.g. on the SEM photographs) is incorrectly assigned to smaller pores. The sum curve of the pore diameter, weighted by area, reveals that 60-80% of the pore space containing pores, which are larger than 100 µm (fig.95). Taken the maximum recorded pore diameter of approximately 100 µm as the upper limit of the “resolution” of the mercury injection method, one becomes a feeling of the induced error.

The cumulative pore size curves, weighted by area, reveal also a small left-shift of the entire pores towards smaller pores with cementation (fig.95). Therefore, the size of pores which control porosity decrease. This goes along with a reduction of the largest detected pore diameter from uncemented samples which have pores up to 1400 µm in diameter (O8) to highly cemented samples which have pore diameter of up to 300 µm (Sample H4).

The pore radii frequency curve, weighted by the volume of mercury injected, reveals also a clear trend of decreasing sizes of the predominant pore radii with cementation, from ~20 µm in the O-series to ~7 - 9 µm in the H-series (fig.97). The "peaks" in the smaller radii range of the cemented samples are seen mainly as artefacts - they are caused by pores connected by much smaller pore throats. The O-series shows much smoother curves with one maximal peak at large pores. Nevertheless, pore radii as small as 6 µm are also detected. The G-series reveals a very heterogeneous pore distribution with either more small pores or more likely, more pores which are connected through smaller pore throats. The samples of the H-series appear to have again a more homogeneous peak-like pore distribution.
It is difficult to establish a quantitative relationship between the two measurement techniques. However, the two techniques had some qualitative features in common which can provide an insight into the evolution of the pores during cementation in the case of the Lochaline Sandstone:

- negative skew of the pore distribution, which means that largest pores contribute mainly to porosity - not influenced by cementation
- only small decrease of the predominant, relative well sorted pore size with increasing cementation
- reduction of the largest pore size with increasing cementation

However, one should note that for transport processes it is not of importance the volume of porous zones with a given conductance but the number of occurrence of such zones, since those will control the flow. However, the determination of "effective" radii for flow is based on the shape of the capillary pressure curve, which is a function of the (mercury-) volume intruded.
Fig. 88: **Sample O6**: SEM photographs are showing fractured, spherical detrital quartz grains in a loose packing. The pore cast reveals that the pores are very well interconnected three dimensionally.
Fig.89: **Sample G16:** SEM photographs show an increasing content of quartz overgrowth and a very low amount of illite (pore bridging). The pore cast reveals a more angular (sheet like) network caused by the subhedral quartz overgrowth. The occurrence of illite micro porosity and dissolved quartz surfaces result in extremely fine pores (A).
Fig.90: **Sample H1**: SEM photographs illustrates the last stage of quartz overgrowth which is marked by the formation of polyhedral quartz crystals. The pore cast shows the resulting slot-like pore apertures. In few cases, several grains may become bounded together by the quartz cement (A).
Fig. 91: **Sample H3**: SEM photographs show grains with smooth well formed crystal faces. Occasionally, the underlying subunits of the quartz overgrowth are visible (A). The pore cast reveals rhombohedral slot-like pore apertures with rounded wholes, which marks grain-grain contacts (B).
Fig. 92: **Sample H1:** Pore cast SEM photograph is showing tubular apparent pore aperture marked by curved plastic sheet (A) and a network of thin pores (B), which could indicate secondary micro pores.

Fig. 93: **Sample H1:** SEM photograph suggest pore throats tabular in shape and polygonal pore geometry (A) due to the final stage of quartz overgrowth.
Fig.94: **Sample H1:** SEM photograph shows pore filling of quartz, micro porous developed. Such notches occurred due to dissolution and may enhance secondary porosity (terminology of dissolution indicators after Burley S.D. and Kantorowicz J.D., 1986). Similar structures are also attributed to clay minerals replaced by opal (Thiry et al., 1991).
Fig. 95a: **H-series**: Pore fraction histogram of pores weighted by their occurrence. The plotted area curve indicates to which percentage the pores contributing to the total porosity.
Fig. 95b: **G-series**: Pore fraction histogram of pores weighted by their occurrence. The plotted area curve indicates to which percentage the pores contributing to the total porosity.
Fig. 95c: **O-series**: Pore fraction histogram of pores weighted by their occurrence. The plotted area curve indicates to which percentage the pores contributing to the total porosity.

Fig. 96a: Histogram of the total pore diameter distribution of sample O6 and O8. The pore fractions are logarithmic grouped.
Fig. 96b: Histogram of the total pore diameter distribution of samples H1-H5 and O7. The pore fractions are logarithmic grouped.
Fig. 96c: Histogram of the total pore diameter distribution of the G-series. The pore fractions are logarithmic grouped.
Fig.97: Frequency plots of the pore radii grouped by the H-, G- and O-series. The plots reveal a decrease of the dominant pore throat radius with increasing cementation from the O-series to the G- and H-series.
4. 6. 2. 2 Characteristic Length Measurements

Specific surface
The grainsize controls the total wetted surface. The specific surface, which is the wetted surface normalised by the bulk volume, decreases from the samples of the O-series (5.82 - 7.03 m$^2$/m$^3$; mean: 6.48 m$^2$/m$^3$) to the G-series (1.04 - 4.75 m$^2$/m$^3$; mean: 3.39 m$^2$/m$^3$) and H-series (2.71 - 3.77 m$^2$/m$^3$; mean: 3.13 m$^2$/m$^3$). The slightly broader grain size distribution of the samples of the O-series, particular in the small grain size fraction, could explain the slightly higher values of the specific surface. A weak correlation exists with permeability (fig.85).

Radii at 75% of the SEM pore distribution sum curve weighted by occurrence
This parameter indicates the pore radii of the large pores. As seen in the charts in appendix 8.3, 25 percent of the pore radii of cemented samples of series H are larger than 29.8 µm and 25 percent of pore radii of series G are larger than 40.95 µm. The samples of series O contain much smaller large pores - only 25% are larger than 22.17 µm. No correlation exists with permeability, which suggests that fluid flow is not controlled by the largest pores.
[H-series: 12.5 - 45 µm, mean: 29.8 µm; G-series: 25.75 - 60 µm, mean: 40.95 µm; O-series: 15 – 25.75 µm, mean: 22.17 µm]

Radii at 50% of the SEM pore distribution sum curve weighted by occurrence
The average pore radii increase with cementation from the O-series to the G- and H-series. A reduction in porosity coexisting with the enlargements of the average pore sizes could only be explained by reduction of the absolute number of pores (fig.98).
[H-series: 4.5 – 18.25 µm, mean: 11.75 µm; G-series: 8.5 – 18.25 µm, mean: 15.12 µm; O-series: 5 – 8.5 µm, mean: 6.8 µm]

Radii at 50% of the SEM pore distribution sum curve weighted by pore area
This parameter indicates the average size of the pore body in the samples, which primariliy controls the porosity. The average pore size, which contains 50% of the total pore volume, is reduced by a factor of 0.5 during cementation from ~ 152 µm to 73 µm. A weak trend of decreasing pore radius with decreasing porosity is observed (fig.98). The pores in the highly cemented sample, which control porosity, are one order of magnitude larger than the “effective” pores for transport.
[H-series: 50 - 100 µm, mean: 73 µm; G-series: 50 - 100 µm, mean: 73.33 µm; O-series: 125 - 182.5 µm, mean: 152.5 µm]
Effective pore radii for fluid flow

The “effective” radii measurements are closely associated with the pore throat size distribution. It was calculated the threshold radii at the apex (Pittman, 1992), the pore radius related to the entry-pressure (Katz and Thompson, 1987) and the average pore throat radius (see fig.5). It is observed that the average pore radii are always between the radii related to the apex and the pore entry radii (fig.100). From this observation, one has to conclude that it is the dominant mean pore radii, which controls the transport processes.

The effective radii plotted versus porosity shows a satisfying correlation between the two parameters (correlation coefficient = 0.8). The effective radii as defined by Pittman and by Katz and Thompson reveals a decreasing trend with decreasing porosity. Infinite small radii of both trends approach zero at ~2% porosity (fig.99). This “threshold” porosity value of infinite small effective radii correlates very well with the percolation threshold obtained from plot of tortuosity against porosity (fig.79). However, plots of the effective radii versus permeability (fig.78) reveals that only in the high permeable samples (>100 mD) exists a relation between the two parameters.

- Threshold radii related to the apex
  In all plots of the ratio mercury saturation/pressure versus mercury saturation is the apex very well developed and the radius was accurately determined. One should note that the saturation between plateau and steep rise in capillary pressure (threshold pressure) corresponds to the saturation related to the well developed apex (appendix 8.3)
  [H-series: 4.49 - 8.79 µm, mean: 6.71 µm; G-series: 6.87 - 12.01 µm, mean: 9.71 µm; O-series: 13.21 -17.55 µm, mean: 15.78 µm]

- Radii related to the pore entry pressure

- Arithmetic mean of the mercury pore radii
Results

Fig. 99: Plot of porosity versus effective pore radii.

Fig. 100: Bar diagram of the effective radii determined by mercury capillary pressure curve.
Pore body - pore throat ratio
The degree of constructivity, which could be expressed by the ratio of pore body to pore throat size, increases significantly in the low porous samples of the H-series (fig.101). The higher complexity of constructivity with increasing cementation could be caused by smaller pore throat constrictions or by oversized pores. The tortuosity, measured at 1kHz, increases also strongly with decreasing porosity. This is in agreement with Kuhlenkampff’s (1994) suggestion that the electric conductivity is strongly influenced by the constructivity of the pore space at low frequency measurements. However, a plot of the two parameters, constructivity versus tortuosity, revealed a poor correlation (fig.101).

[H-series: 4.6 – 8.7, mean: 7.56; G-series: 3.2 - 5.34, mean: 3.85; O-series: 4.58 - 8.76, mean: 6.28].

Residual percentage of mercury after drainage
Since trapping of mercury is extensive for pore systems with large pore body - pore throat ratios (Wardlaw, 1980), the parameter reflects also the complexity of the constructivity of the pore system. It is observed an increase of the residual mercury saturation with decreasing porosity (fig.101). The massively decrease in recovery efficiency in the highly cemented samples indicates a significant change in pore structure – the pore system consists of larger pore bodies or/and smaller pore throat constrictions. One should note that the residual mercury saturation correlates well with the increasing tortuosity of the pore system.

[H-series: 34 – 71, mean: 47.6; G-series: 3 - 43, mean: 15.2; O-series: 10 - 17, mean: 14.8]

Lower length limit of the fractal distribution of pores
Fractal pores are associated with power law behaviour of the pore widths at small length scales. Euclidian porosity is shown by a lack of power law behaviour and by the appearance of a peak in the pore volume (respectively area) distribution at the long-length scales. All samples had both, fractal and Euclidian pores. The physical properties of the two types is expected to be different. However, one should also note that all “effective” pore radii for transport were Euclidean pores. Experimental measurements have shown that grain/pore interfaces in most sandstones can exhibit fractal behaviour from $10^{-2}$ µm to $10^{2}$ µm (Thompson et al., 1987; Wong et al., 1986; Hansen and Skjeltorp, 1988; Krohn, 1988). This is in agreement with the following results:

- SEM pore area distribution: The pore diameter distribution behaves fractal within the length limits of ~20 to 100 µm at the chosen SEM - image magnification. The upper length limit is much smaller than the predominant grain size.
- Mercury pore throat volume distribution: The lower limit of the fractal behaviour of the pore radii distribution is thought to reflect the resolution limit of the technique employed. The lower length limit is reached at pores radii of around 0.1 micron in the case of the mercury method. However, it varies with porosity (fig.102). Since the upper and lower
length limit shows an approximately constant spacing, it is though that the length scale is a function of the sample size. The upper length limit is always smaller than $R_{\text{apex}}$.

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**Fig.101**: Plots of pore body / throat ratio pore radii and residual Hg-saturation versus $\tau$ and $\chi$. The residual Hg-saturation correlates better with $\tau$ and $\chi$.

**Fig.102**: Lower and upper length limit of the fractal mercury pore radii distribution.
4. 6. 2. 3 Fractal Dimension

The fractal dimension was determined by using three methods: mercury injection (Friesen and Mikula, 1987), scatter electron microscopy (Krohn, 1988) and measurements of dielectric properties (Wong et al., 1986; Ruffet et al., 1991; LeMehaute and Crepy, 1983). It is established that the pore-grain interfaces are in most sedimentary rocks are ideal fractal (Thompson, 1987) whereas the pore volume must not have the same fractal dimension (Wong et al., 1986 and Hansen and Skjeltorp, 1988). Pape et al. (1987a) assumed also that the ratio of pore volume to total volume is a non-ideal fractal accordingly to Rigaut (1984), with respect to measuring as it converges against a finite limited maximal porosity, but behaves as an ideal fractal, which goes to zero, during compaction (Pape et al., 1987b).

It is purposed that fractal dimension is closely related to geometrical properties of a fractal interface, such as the roughness of the pore / grain interface. The surface of the grains in the H-series are massively altered due to cementation, which incorporates smoothing of the grain surfaces as seen on SEM photographs (fig.91). Therefore, it was expected that the fractal dimension decrease with decreasing porosity. Such a physical behavior was observed in plotting the fractal dimensions obtained by the models of LeMehaute (D: 2.5 -> 2) and Ruffet (D: 3 - 2.3) against porosity (fig.103). However, the fractal dimension derived from mercury injection and the model of Wong show a continuously increasing trend of the fractal dimension with decreasing porosity, the fractal dimension derived from SEM-image analysis exhibited no clear trend.

One should note that highly permeable samples (> 100mD) showed no fractal structure, when the dielectric property models were employed. Fractal structures were detected, in all samples examined by SEM and Hg-injection. Fractal dimensions calculated from mercury injection and SEM-image analysis represented realistic values between 2.1 and 2.9.

It is thought that the fractal dimension derived from mercury injection is related to the fractal pore volume and not only to the roughness of the grain-pore interface. For this purpose it was determined the fractal porosity. The Euclidean part of the pore-space was thought to represent the unaltered porosity whereas the ratio of relative porosities associated with the fractal regimes, would be a measure stick of the diagenetic alteration of the pore space. The fractal porosities were ranging from ~0.4 to 3.4%. Although the dimension was independant of the fractal porosity, the fractal dimension correlated well with the ratio fractal/total mercury porosity (fig.104). A crossplot reveals marked higher dimensions (> 2.6) of samples with high ratio’s (> 0.2). The relative measure of fractal porosity versus total porosity acted as a measure stick of the degree of alteration in agreement with Aharonov’s (1997) results - the higher the ratio (\( \frac{\text{Frac}}{\text{Tot}} \)), the higher the degree of cementation (fig.104).

Since the fractal porosity is thought to contribute not to transport, it was examined whether the Euclidian part of the porosity correlates better to permeability. Indeed, the Euclidean mercury porosity showed a slightly better correlation with permeability than the total porosity (fig.77).
Fig. 103: Plot of fractal dimensions derived from different methods versus permeability. The fractal dimension calculated from mercury injection and electric measurements (Wong’s model) is inverse related to permeability.

Fig. 104: The fractal dimension shows increase with the diagenetic measure fractal / total Hg.
(after Aharonov, 1997).
V. Discussion

Pore structure / poroperm trend

In the Lochaline Sandstone samples exist an excellent correlation between porosity and permeability. This correlation is expected because the sandstone has a nearly constant grain size. Both parameters are massively affected by a single factor: cementation. Compaction effects are neglected, since the compaction affects the whole formation to approximately the same degree and since in well sorted rocks both compaction and overgrowth have virtually the same effect on permeability as a function of porosity (Bryant and Blunt, 1992). The cementation affects the permeability through three processes:

- the absolute amount of pores decreases, hence the porosity decreases
  - samples G13 and H3 reveal even the same skewed shape of the pore distribution as the uncemeted samples of the O-series, suggesting a proportional-equal decrease of the large and the small pores.
- the arithmetic averaged pore size decreases, which contains 50% of the pore space
  - the process of cementation leads predominantly to homogenisation causing elimination of smaller pores and decrease of larger pores
- both "effective" pore radii decrease - the radii related to the apex as well as the radii related to the entry pressure

Those changes in the pore structure cause a decrease in permeability over several orders of magnitude from 2000 to 1 milliDarcy. However, it exhibit not a linear relationship between log K and \( \phi \) as determined in other relatively pure quartz sandstone formations as the Cretaceous Houston sandstone (Thompson, 1978) or the Tertiary Buntsandstein (Füchtbauer, 1967). The entire poroperm trend in the Lochaline Sandstone relates permeability to a power 3.8. Better correlations in the low porous samples are obtained by grouping the data into two families, for which distinctly different poroperm relationships can be identified: for \( \phi > 12\% \) the permeability data are in good agreement with the power law with an exponent of 1.6. In contrast, the regime for \( \phi < 12\% \), requires porosity raised to a power of 5.5 to fit the data trend. The correlation law shows for both regimes the same mathematical shape, but not the same characteristic parameters. Finally it is also suggested a percolation threshold at \( \phi \sim 2\% \). This suggestion is based on the exponential increase of tortuosity in the very low porous samples. The poroperm trend shows similar patterns in comparison to the more extensive studied and textural similar Fontainebleau Sands:

Fontainebleau Sandstone:

- One power law: \( \phi^{3.8} \) (Doyen, 1988), \( [\phi-2.5]^3 \) (Mavko and Nur, 1997)
- Two regimes: high \( \phi \) (9-28%): 0.303\( \phi^{3.05} \), low \( \phi \) (3-9%): 2.75*10^{-5} \( \phi^{7.33} \) (Bourbie et al., 1985)
- Three regimes: high \( \phi \): \( \phi^3 \), low \( \phi \): \( \phi^{\gamma} \) (the power law no longer applies and an accelerated reduction in permeability was found; the change between the high and the low
porosity trend is defined by the crossover porosity); lowest \( (< 2\%): \) in the third regime becomes permeability to small to be measured, no percolation occurs (Zhu et al., 1995).

Lochaline Sandstone:
- One regime: \( 3.82 \)
- Two regimes: high \( (12>20\%): 12.78 \) \( 1.66 \), low \( (4<12\%): 7*10^{-4} \) \( 5.59 \), it exists a percolation threshold at \( \sim 2\% \).

The pore geometry is marked by two distinctive structural styles in the two poroperm regimes. The low porous samples consist of highly altered primary porosity, extensively occluded with quartz overgrowths, and coupled with significantly secondary pore development. Primary porosity is reduced to flat, slot-like pores along the boundaries of adjoining quartz overgrowth. Although most of the porosity occurs in the secondary pores, the narrow slots often provide the only flow paths and create a “bottleneck” for movement of pore fluids. This pore structure in the low porous samples is comparable with the pore structure of tight gas sand reservoirs like the Cretaceous Travis Peak formation (Soeder and Chowdiah, 1990). In the high porous samples, which represent the bulk of the Lochaline Sandstone, spherical grains control the pores.

The ability of porosity in the prediction of permeability appears to be controlled by the relation of the effective radii to porosity. In the low porous sample, porosity is related by a very high power of 5 to permeability because large pores, which are not contributing to flow, mainly control porosity. It is thought that the percentage of the ineffective secondary dissolution porosity increases in the low porous samples. This leads to an accelerated reduction in permeability with the measured porosity. Whether this process results in stabilisation of the mean pore size as suggested by Zhu’s (1995) referring to data of the Fontainebleau sands (Fredrich et al., 1993) could not be confirmed, since the scatter of the mean pore size data are too large.

However, in the case of the LSst, reduction in the amount of pores (not the size of pores) results not only in reduced pore space, but also in a higher tortuosity of the flow path. Additionally to the resulting longer pathway, the higher tortuosity is also caused by a more complex pore constrictivity. This situation implies that a smaller percentage of total porosity is part of the transport process in the altered pore system.

The high exponent of the poroperm trend of cemented samples (red line) in comparison to the superimposed uncemented poroperm trend (black line in sketch) probably reflects a marked continuous increase in additionally ineffective porosity. Ineffective porosity includes unconnected porosity, dead end porosity and porosity which is not related to the effective pore radii (e.g. dissolution porosity, fractal porosity).
The primary affection of the cementation is suggested to be conductivity loss, with pore size reduction being of secondary importance. However, it is mainly the contrasting path of the evolution of pore bodies and pore throats which determines the change in transport properties with decreasing porosity. Similar conclusions were reached by numerical studies of Wong et al. (1984) and by an empirical case study of the Fontainebleau sands (Doyen, 1988).

One should note that the electrical conductance is related to porosity by a single power law of 1.88. The trend appears to be not significantly affected by the different styles in microstructure. Such low m-values in comparison to the m-value of 2 as suggested by Archie are not unusual since Carothers (1958) and Timur et al. (1972) obtained m-values of sandstone from FF versus φ plots which are even lower than 1.8. For the comparable pure quartz Fontainebleau Sandstone, Doyen (1988) determined the m-value to be 3.7 for φ<10% and a value of 1.9 for φ>10%. Adler et al. (1992) generated a contradictory low m-value of 1.62 for the same sandstones from regression analyses on data presented by Jacquin (1964).
Silification
- Conditions for silica precipitation / origin of the sub-basaltic hard bands?

Generally, such very clean sands are mostly common in shallow marine, high energy ridge margins and central ridge facies (Tillman, 1985; Tillman and Martinson, 1985). They are commonly considered to be derived from preexisting sandstones and so are of multicircle origin (Suttner et al., 1981). Regional settings (being overlain by calcereous lithologies) shows that the LSst was submerged relatively rapidly by continued marine transgression. However, prior to the extrusion of basalts (gap of ~ 40 million years) that overlie the LSst, a period of subaerial exposure occured. During this period, it is likely that the formation was altered by leaching and cementation.

The cemented horizons within the Lochaline Sandstone are suggested to be typical silcretes as defined by Summerfield (1983): “silcrete is an indurated product of surfacial and near surfacial silification, formed by cementation and/or replacement of bedrock, weathering deposits, unconsolidated sediments, soil or other materials and produced by low temperature physico-chemical processes and not by metamorphic, volcanic, plutonic, or moderate to deep burial diagenetic processes”. The Lochaline formation do not contain significant siliceous biota nor do they exhibit significant pressure solution features; so biogenic processes and pressure solution as sources of silica are ruled out. In general, the nature of the silica phase reflects the chemistry of the solution from which it precipitated. Solutions with relative low concentrations of silica and low levels of impurity ions can precipitate quartz overgrowths on skeleton grains (Williams and Crerar, 1985; Delmas et al., 1982).

Predominantly such crystallographic continuity overgrowth occurs in the case of the Lochaline Sandstone, which is characteristically for the “quartzitic type of silcrete” after Smale (1973). In this type, the typical clayey sub-silcrete profile is not generally present, as orthoquartzite is not susceptible to alteration. Nevertheless, antimony (Sb) was found as the most abundant tracer element. It is typically enriched in secondary iron oxides due to weathering processes under arid oxidising conditions (Götz and Lewis, 1994). Such oxidising postsedimentary conditions are causing also the observed changes in the mineralogy, in the <2 µm and 2<20 µm fractions after Götze and Lewis (1994). The observed secondary anatas are also indicate a weathering profile (Summerfield, 1983). Bleaching and oversized pores are further evidence of an altered sand profile. This type of secondary porosity is mainly produced due to leaching by acid meteoric water after Shanmugam and Higgins (1988) and Mathisen (1984).

In many low temperature systems, silica precipitates from natural aqueous solutions as amorphous silica. The suggested diagenetic sequence follows the following transformation: opal-A (amorphous silica) -> opal-CT -> quartz. (Williams and Crerar, 1985). However, neither amorphous silica was observed in thin sections nor in the XRD-analysis. The solubility of the different phases of silica is close related to this order where opal-A has the
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highest solubility of 60 - 300 ppm, cristobalite \(~20-30\) ppm and quartz \(~6-10\) ppm, all measured at 25°C (Iler, 1979). It is therefore possible that amorphous silica was precipitated and recrystallised during diagenesis producing a silicious cement to the grains. The development of horizontal “layers” of caves associated with the cemented horizons (e.g. at outcrop 5) could be interpreted as relics of dissolved amorphous nodules as reported from pedogenic silcretes (Thiry and Miles, 1991). However, the caves could also be interpreted as recent surface weathering structures, like “solution basin” as described by Wray (1997). It is more likely that it was a dilute silica solution from which only quartz was directly precipitated. Rather few quantitative data exists for precipitation at low temperature conditions and many are conducted in deionised water (Dove, 1995). Iler (1955) showed that it is possible to crystallise quartz from solutions of silica below equilibrium concentration. Mackenzie and Gees (1971) have crystallised quartz overgrowth from seawater solutions of silica of 4.4 \(\pm 0.3\) ppm at 20°C and 1atm and with a pH of 8.1.

The observed apparent sesquioxide coating on the grains (dust lines) may have been influential in the crystallisation of quartz, since the solubility may be reduced by absorption of silica on to iron and aluminium oxides (Jones and Handreck, 1963). Harder (1971) showed that amorphous hydroxides of Al and Fe are capable of removing silica from very dilute solutions at low temperatures (3-20°C).

Up to about pH 9 the silica is largely undissociated, but at a pH greater than 9 dissociation and polymerisation occurs and \(\text{H}_3\text{SiO}_4\) becomes predominant. This is reflected in the increase in solubility in highly alkaline solutions. Berger et al. (1994) found that electrolytes increase dissolution rates at 25°C and pH 6.5 - 7 by a factor of 6. Additionally to a decrease in pH, a solution becomes also supersaturated with respect to silica if there occurs a decrease in temperature or volume of fluid (e.g. by evaporation). The same mechanisms are suggested to be primary relevant for formation of silcrete (Summerfield, 1983). But since temperature fluctuations are certainly far more rapid than the rate of reaction of silica to supersaturation and one also consider the relatively slow rate of change in solubility at temperatures near the surface, temperature drop is unlikely to account as a factor for silcrete genesis. However, field observations (McCarthy and Ellery, 1995) in an arid flood plain environment suggest that a drop in pH in groundwater play little role in silica precipitation. Important is the loss of water, which increases the salinity of the groundwater, inducing silica precipitation. Ancient silcrete are formed by rising silica charged solutions meeting percolating solutions containing NaCl, Fe\(_2\)O\(_3\), Al\(_2\)O\(_3\) or of lower pH accordingly to Smale (1973).

The presence of marked lateral and vertical variations in of silcretes at Beinn na h-Uamha probably, even within single outcrops, reflects major water table fluctuations with periods of percolation during low water periods. Columnar structures are observed at the outcrops, mainly in the upper part of the sequence. Such structures are also recorded from Australian silcretes and suggested to be formed by upward-percolation of siliceous water (Smale, 1973).
and Hutton et al., 1972), by downward percolation (Thiry and Milnes, 1991) and/or by
groundwater silification (Simon-Coincon et al., 1996). Wray (1997) suggested that such
structures are not only generated by solutional attack in the subsurface but formed also
subaerially as drainage channels by dissolution (defined as rinnen or rundkarren).
The observed structures sheet-like precipitation horizons of silica within the Lochaline mine
are vertical less variable and lateral extensive. This may indicate only minor fluctuations in
water table caused by a different environment.
It is obvious that both erosion and deposition must be minimal at both localities so that a
sufficient period of surface stability can occur for silcrete to form. The nearly horizontal
cemented layer also suggests a flat relief of the surface. A semi-arid environment is also
required, with sufficient spasmodic rainfall to cause substantial fluctuations in water table. No
field evidence supports that the water table fluctuations are controlled by a progressive
dissection of the landscape as suggested by Thiry (1988) for the similar silcrete of the
Fontainebleau Sandstone. Detrital grains of the cemented horizons are examined to be better
sorted than the uncemented grains. This could indicate that cementation is related to higher
permeable zones, preferred by konvective flow. Surface accumulation by evaporation of
siliceous solution may also take part as indicated by clasts of silified chalk in the
paracoglomerates of the Turonian Laig Gorge Sandstone (Braley, 1990) and observed erosive
surfaces between cemented and uncemented horizons of the LSst. Both, modern surface and
subsurface silification processes are observed in arid environment like the Okavango flood
plain (McCarthy and Ellery, 1995).
A further possible process could be the precipitation of quartz at the seawater – sediment
interface: sufficient quartz dissolved in seawater during bedload abrasion of detrital quartz
grains and subsequent precipitated below the sediment-water interface. Three reports of
quartz overgrowth forming exists in the following modern marin subaqueous environments:
beach sands (Dapples, 1967), tital channel sands (Baltzer and Le Ribault, 1971) and shelf
sands (Fillon et al., 1978). However, it is unlikely that the observed thick cement
accumulations are formed by such a process.
It has also been suggested for sub-basaltic silcretes that basalts are genetically related to the
silcretes (Brown, 1972), e.g. they provide heat, causing quartz in the underlying quartz rich
sediments to dissolve, and silica to precipitate on cooling. Hexagonal columns, similar to
those exposed at outcrop 2, have been associated with lava flow and are interpreted as formed
due to heating and desiccation (Judd, 1878). However, such a direct influence of the overlying
basalts can be ruled out: it is observed a more complex history of the cement overgrowth than
can be accounted for by a single thermal event. Neither it is suggested that the basalt acts as
source of silica e.g. through weathering and percolation of meteoric water, because it exists no
tracer element correlation between the LSst and the basalt. The observed insignificance of the
silcrete-basalt association is in agreement with results of similar sub-basaltic silcretes in
Australia (Taylor and Smith, 1975).
VI. Summary

- Lochaline Sandstone is ideal for use in testing models dealing with natural porous media because of
  - its high purity
  - it has two distinct microstructural styles

- “Effective” radii models tend to underestimate permeability.

- The pore geometry has a wide spectrum ranging from nearly elongate-subspherical to slot-like or tabular pores.

- Permeability is ranging over several orders of magnitude: from ~ 1mD to 2300 mD.

- Helium porosity values range continuously from 4.1% to 20.9%.

- The percolation threshold is reached at ~ 2% porosity.

- Northwards, the cementation horizons are lateral less extensive and vertical more variable which probably reflects different environments during genesis.

- The overlying basalt have no significance in genesis of the silified horizons.

- Porosity measured at different scales with different techniques are not identical.

- The grain size distribution derived from mercury injection massively overestimates the actual grain size.
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VIII. Appendices

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