nagra
National Cooperative for
the Disposal of Radioactive Waste

TECHNICAL REPORT 91-16

IMPLICATIONS OF LONG-TERM TRANSIENT FLOW, COUPLED FLOW AND BOREHOLE EFFECTS ON HYDROGEOLOGICAL TESTING IN THE OPALINUS CLAY: PRELIMINARY STUDY WITH SCOPING CALCULATIONS

S.T. HORSEMAN
J. ALEXANDER
D.C. HOLMES

DECEMBER 1991

Fluid Processes Research Group
British Geological Survey
Keyworth, Nottingham, NG 12 5GG
England

Hardstrasse 73, CH-5430 Wettingen/Switzerland, Telephone +41-56-37 11 11

nagra National Cooperative for the Disposal of Radioactive Waste

TECHNICAL REPORT 91-16

IMPLICATIONS OF LONG-TERM TRANSIENT FLOW, COUPLED FLOW AND BOREHOLE EFFECTS ON HYDROGEOLOGICAL TESTING IN THE OPALINUS CLAY: PRELIMINARY STUDY WITH SCOPING CALCULATIONS

S.T. HORSEMAN
J. ALEXANDER
D.C. HOLMES

DECEMBER 1991

Fluid Processes Research Group
British Geological Survey
Keyworth, Nottingham, NG 12 5GG
England

Hardstrasse 73, CH-5430 Wettingen/Switzerland, Telephone +41-56-37 11 11

This report was prepared as an account of work sponsored by Nagra. The viewpoints presented and conclusions reached are those of the author(s) and do not necessarily represent those of Nagra.

RESUME

Au cours d'une réunion entre la CÉDRA et les représentants de BGS, le 24 mai 1989 à Keyworth, on a examiné un certain nombre de processus pouvant être responsables des pressions hydrauliques anormalement basses observées lors d'essais hydrauliques en forages dans l' "Argile à opalinum" (marnes aaléniennes). Une pression hydraulique anormale fut définie comme une pression ne pouvant être interprétée en termes d'écoulement gravitationnel normal en régime permanent.

En examinant le problème, il apparut que trois types de processus pourraient être invoqués; le premier est associé à un écoulement transitoire à long terme, le second à un écoulement couplé (en particulier avec les forces osmotiques), et le troisième à des effets de forage comme le gonflement, la perturbation des pressions in situ et la déformation plastique des parois du forage.

Ce rapport représente une étude préliminaire des processus identifiés. L'approche choisie a été d'examiner chaque type de processus individuellement, dans un premier temps à un niveau général, et ensuite avec des références spécifiques à l'Argile à opalinum. Des modèles théoriques simples ont été établis et un grand nombre de calculs a été effectué dans le but de distinguer les effets d'ordre primaire ou secondaire. Les paramètres de base hydrogéologiques, géotechniques et géochimiques de l'argile litée sont obtenus à partir des données d'analyses et d'essais antérieurs, ou sont calculés à partir des relations connues entre ces paramètres. Les paramètres pour lesquels aucune donnée n'existe ont été tirés de la littérature sur les argiles litées.

Ce qui ressort de cette étude est que, bien que les mécanismes de mouvement de l'eau souterraine dans les roches argileuses soient pour le moment mal connus, certains processus pourraient avoir des effets hydrogéologiques assez importants pour masquer complètement les réponses normales des essais hydrauliques. Ceci peut avoir des répercussions sur le développement d'une stratégie pour la caractérisation d'une site, en ce qui concerne la modélisation et les analyses de sécurité.

Nos calculs suggèrent qu'un écoulement transitoire à long terme dans l'Argile à opalinum peut être la conséquence de la décompression des roches associée à l'érosion des couches de surface. Ce processus sera le plus actif là où l'érosion a été la plus forte (p. ex. au tunnel du Wisenberg et dans la vallée de Homburg). De plus, des

déformations néotectoniques étant postulées dans plusieurs régions de Suisse, y compris dans le Jura, il est possible qu'une déformation tectonique des argiles influence le régime hydraulique dans ces dernières.

L'image générale qui ressort de nos calculs concernant le mécanisme de surconsolidation est que les écoulements de fissure se rééquilibrent assez rapidement après exhumation, mais le réajustement des teneurs en eau dans la matrice rocheuse occupe une bien plus longue période. Il est retardé par la cohésion diagénétique des minéraux argileux et la formation de ciments. Ainsi, les écoulements de fissure pourraient être considérés comme essentiellement déphasés par rapport aux écoulements dans l'argile intacte (écoulement de matrice).

La faible porosité et la généralement forte teneur en argile de l'Argile à opalinum suggèrent que cette dernière pourrait également agir comme une membrane semi-perméable avec un flux osmotique non seulement aux limites des couches mais aussi à l'intérieur de celles-ci. Nos calculs montrent que l'osmose peut avoir des effets importants sur les mesures de pression hydraulique dans les roches argileuses. Ces effets doivent être pris en compte lors d'essais hydrauliques dans de telles roches. Un écoulement couplé peut aussi être très important lorsqu'on modélise les mouvements de l'eau souterraine en roches argileuses.

Si l'on admet que le flux osmotique est une composante importante du flux total en roche argileuse, alors le comportement transitoire à long terme doit être considéré comme une réponse à un déséquilibre hydraulique et chimique.

Un profond enfouissement suivi d'exhumation de l'Argile à opalinum ont engendré dans celle-ci une nette "soif" d'eau. Cette tendance à avaler de l'eau peut être interprétée en simples termes mécaniques comme la conséquence de l'expansion après surconsolidation, ou en termes chimiques comme un phénomène de succion associé à l'osmose et/ou à l'hydratation opérant à échelle microscopique. De puissantes succions (pressions interstitielles négatives) peuvent se développer dans les roches proches de la surface. De telles succions sont tout à fait compatibles avec la forte capacité de gonflement observée dans l'argile. Ceci joue un rôle fondamental dans nos discussions concernant les écoulements transitoires à long terme et les écoulements osmotiques (couplés) dans les roches argileuses.

Ainsi nous voyons que dans les roches argileuses, particulièrement dans les argiles litées très compactes comme l'Argile à opalinum, il y a un nombre considérable de raisons pour lesquelles la pression hydraulique (déterminée de façon habituelle) pourrait ne pas être interprétable en termes d'écoulement gravitationnel normal en régime permanent. En fait, dans beaucoup de milieux argileux, il est difficile d'imaginer le cas d'une pression qui ne serait pas "anormale".

Les effets de forage, associés aux modifications mécaniques, thermiques et chimiques des roches en cours de forage et d'essai, peuvent avoir une influence significative sur la réponse hydraulique mesurée pendant les essais. Bien que le gonflement dû à l'introduction d'eau fraîche est probablement le facteur principal, la perturbation in situ du champ des contraintes et la déformation plastique de la roche peuvent avoir dans les forages profonds un effet considérable sur la distribution des pressions intersticielles autour du forage. Si le dispositif de mesure est caractérisé par un faible temps d'ajustement ("compliance"), les mesures peuvent aussi être affectées par la fermeture progressive du trou.

Si nous examinons le cas particulier de charge hydraulique anormale dans le forage RB26B, sur le traçé proposé pour le tunnel du Wisenberg dans la vallée de Homburg (cf. section 6.2), nous ne pouvons exclure totalement de très simples explications, comme la présence d'air dans les intervalles de test. Cependant, les pressions anormales peuvent s'expliquer (semi-quantitativement) en admettant que la pression totale **h** en chaque point de ce forage s'exprime par

$$\mathbf{h} = z + h_{pp} + h_{sp}$$

où z est la pression due à l'altitude, h_{pp} la pression dans les macropores (liée à la pression hydrostatique) et h_{sp} la pression du soluté dans les macropores. Si ce scénario est correct, alors il a de très importantes conséquences pour la planification et l'interprétation des essais hydrauliques dans les roches argileuses. La conséquence la plus importante est que la pression hydraulique déterminée par de tels essais dépend de la concentration chimique du fluide de test. Ceci remet en question l'usage habituel d'eau déionisée comme fluide de test. Des liquides non polarisés et non réactifs pourraient être plus appropriés. D'autre part la nature chimique des fluides de test devrait être considérée comme une variable dans le développement de méthodologies d'essais hydrauliques dans les roches argileuses.

Il ressort de cette étude une relation intéressante entre le flux chimico-osmotique dans la roche argileuse (flux couplé) et le processus physico-chimique du gonflement inter-particulaire. A proximité d'un forage, les deux processus sont pratiquement inséparables. Ils relèvent tous deux d'un mécanisme par lequel l'eau est transférée de l'intervalle de test au milieu rocheux, et ils sont tous deux sensibles à la nature chimique des fluides de test. Ceci suggère que ces deux processus pourraient être décrits par une théorie unifiée. Ce n'est probablement pas un hasard si les caractéristiques de l'argile qui font d'elle une membrane osmotique efficace sont celles là mêmes qui la dotent d'une grande capacité de gonflement.

Les conclusions apportées par ce rapport sont si importantes pour la caractérisation des sites hydrogéologiques en milieu argileux que des études supplémentaires in situ et en laboratoire s'imposent. De telles études devraient être appuyées par le développement parallèle des modèles théoriques. Nous distinguons trois axes prioritaires: (a) le développement d'une méthodologie d'essais hydrauliques en forage spécifique aux roches argileuses associé au développement de modèles théoriques nécéssaires au traitement des données; (b) le développement d'un modèle numérique relativement simple, si possible uni-dimensionnel, permettant des analyses de sensibilité des écoulements transitoires sous gradients hydrauliques et chimiques; et (c) l'étude plus approfondie des conséquences d'une déformation tectonique sur l'hydrogéologie régionale de l'Argile à opalinum. Des recommandations spécifiques sont presentées dans le chapitre 9.

ZUSAMMENFASSUNG

Anlässlich einer Besprechung vom 24. Mai 1989 in Keyworth (England) wurden zwischen Vertretern der Nagra und dem British Geological Survey (BGS) mögliche Ursachen für die anomal niedrigen hydraulischen Druckhöhen diskutiert, die in Bohrungen im Opalinus-Ton beobachtet worden sind. Solche anomalen Druckhöhen können nicht mit den üblichen Gesetzen für gravitativ induzierte, advektive Strömung unter stationären Bedingungen interpretiert werden.

Als Ursachen für diese anomalen Druckhohen schienen folgende Erklärungen denkbar: (1) langzeitig transiente Fliessbedingungen, (2) gekoppelte Strömung, vor allem mit osmotischen Prozessen und (3) Bohrlocheffekte, zum Beispiel Quellung,Veränderungen

der herrschenden Gebirgsspannungen und plastische Deformation der Bohrlochwände.

Dieser Bericht stellt eine vorläufige Abklärung dieser drei identifizierten Prozesse dar. Jeder wird für sich betrachtet, erst auf allgemeiner Stufe, dann spezifisch im Zusammenhang mit dem Opalinus-Ton. Innerhalb eines einfachen theoretischen Rahmens werden verschiedene abklärende Vorberechnungen durchgeführt, um die wichtigsten Auswirkungen von Nebeneffekten unterscheiden zu können. Grundlegende hydrogeologische, geotechnische und geochemische Parameter für den Opalinus-Ton basieren auf vorhandenen Testdaten, oder werden aus bekannten Parametern und Zusammenhängen errechnet. Wo keine Daten vorhanden sind, werden charakteristische Eigenschaften aus der Literatur übernommen.

Obwohl zur Zeit Ungewissheit herrscht über die genauen Mechanismen der Grundwasserströmung in tonigen Gesteinen, scheinen einige Prozesse tiefgreifenden Einfluss auf die Hydrogeologie zu haben. Die Bedeutung dieser Prozesse geht weit über die Interpretation von Bohrlochdaten hinaus; es sind Auswirkungen auf mögliche Strategien zur Standortcharakterisierung, auf die Modellierung, sowie auf die Sicherheitsanalysen zu erwarten.

Aufgrund der rechnerischen Abschätzungen scheinen im Opalinus-Ton langfristig transiente Strömungen plausibel zu sein. Diese sind mit Spannungsänderungen und Deformation des Gesteins nach der ursprünglichen Überlagerung und der nachfolgenden Druckentlastung während der Erosion darüberliegender Schichten verknüpft. Aufgrund der Häufigkeit,Verteilung und Activität vieler neotektonischer Störzonen, kann zusätzlich in vielen Teilen der Schweiz damit gerechnet werden, dass sich vorab in niedrig permeablen Gesteinen wie dem Opalinus-Ton, infolge von Spannungs- oder Verformungs-induzierten Änderungen des Porendruckes anomale hydrogeologische Verhaltnisse bilden.

Beim Mechanismus der Druckentlastung ergibt sich aufgrund der rechnerischen Abschätzungen folgendes generelles Bild: die Grundwasserströmung in Klüften gleicht sich relativ rasch an die neuen Bedingungen an, hingegen verändern sich die Druckverhältnisse im intakten, ungeklüfteten Tongestein sehr viel langsamer. Im letzteren Fall scheint der Grad der diagenetischen Kompaktion (Bindung zwischen Tonmineralien, Zementierung, usw.) der ursprünglich plastischen Tonmineralmatrix eine gewisse Rolle zu spielen. Im allgemeinen Fall ist deshalb zu erwarten, dass die

Wasserströmung in Klüften kaum je in Phase sein wird mit der Grundwasserbewegung im intakten Tongestein (Matrix-Strömung).

Die niedrige Porosität und der hohe Gehalt an Tonmineralien lassen den Opalinus-Ton als wirksame semipermeable Membrane deuten, die sowohl innerhalb,wie auch über die Tonschichten hinweg, osmotisch getriebene Wasserbewegungen zulassen könnte. Rechnungen ergeben einen starken Einfluss der osmotischen Drücke auf Messungen von hydraulischen Druckhöhen in Bohrungen. Beim hydraulischen Testen in Tongesteinen müssen solche Effekte in Betracht gezogen werden. Gekoppelte Strömung mag beim Modellieren der Grundwasserströmung in Tongesteinen ebenfalls von wichtiger Bedeutung sein.

Falls osmotische Prozesse eine wesentliche, treibende Kraft für die Grundwasserbewegung in tonigen Gesteinen sind, dann muss das langfristig transiente Verhalten als das Zusammenwirken hydraulischer und chemischer Ungleichgewichte betrachtet werden.

Die ursprünglich hohe Überlagerung und die nachfolgende Druckentlastung (durch Erosion der überliegenden Sedimente) verursachten einen ausgeprägten "Durst" des Opalinus-Tons. Diese Tendenz, Wasser aufzusaugen, kann man sich vereinfacht folgendermassen vorstellen: entweder mechanisch, als langsame Erholung von einer ursprünglichen Drucküberlastung, oder chemisch, wobei Saugspannungen als Folge von osmotischen Prozessen oder Hydratisierung auf mikroskopischer Skala entstehen können. Grosse Saugspannungen, d. h. negative Porendrücke, entstehen deshalb vorwiegend in oberflächennahen Schichten. Das Vorkommen solcher Saugspannungen ist besonders plausibel im Lichte des beobachteten Quellvermögens des Opalinus-Tons. Diese Saugspannungen sind von grundlegender Bedeutung sowohl bei der Diskussion langzeitiger Transienten wie auch bei der Interpretation einer osmotisch induzierten, gekoppelten Strömung im Tongestein.

Es bestehen eine Reihe von Gründen , weshalb in tonigen Gesteinen und vor allem in hochkompaktierten Tonschiefern, wie dem Opalinus-Ton, konventionelle Messungen von hydraulischen Druckhöhen nicht einfach mit den Gesetzen der gravitativ getriebenen Strömung unter stationären Bedingungen erklärt werden können. Es ist durchaus möglich, dass in vielen tonigen Gesteinen, vor allem in der Nähe der Erdoberfläche, im Normalfall mit anomalen Druckhöhen zu rechnen ist.

Bohrlocheffekte, die auf mechanische, thermische und chemische Veränderungen im Zusammenhang mit dem Bohr-oder Testablauf zurückzuführen sind, können einen bedeutenden Einfluss auf das Resultat nach dem hydraulischen Testen haben. Obwohl Quellvorgänge infolge vom Einbringen von Süsswasser wahrscheinlich die wichtigsten Effekte darstellen, können in tiefen Bohrungen Veränderungen des umgebenden Spannungszustandes, und die damit verbundene plastische Deformation des Gesteins, wesentliche Störungen der Porendruck-Verteilung in unmittelbarer Nähe des Bohrlochs hervorrufen. Falls das Testsystem eine niedrige "Compliance" hat, können die Messungen zusätzlich durch zeitabhängiges Verformen der Bohrlochwand verfälscht werden.

Im folgenden wird auf den spezifischen Fall anomaler Druckhöhen-Messungen in der Bohrung RB26B im Gebiet des geplanten Wisenbertunnels im Homburger Tal (siehe Kapitel 6.2) näher eingegangen. Da man das Vorhandensein von Luft in den Testabschnitten nicht mit Sicherheit ausschliessen kann, wäre dies wohl die naheliegende und einfachste Erklärung der Messdaten. Dennoch soll eine semiquantitative Interpretation der anomalen Druckhöhen hergeleitet werden, wobei in jedem Punkt der Bohrung folgende Beziehung gelten soll.

$$\mathbf{h} = z + h_{pp} + h_{sp}$$

z entspricht das geodätischen Potential (elevation head), h_{pp} ist das Druckpotential in den Makroporen (im Zusammenhang mit dem hydrostatischen Porendruck) und h_{sp} ist das chemische Potential in den Makroporen. Falls diese Beziehung gilt, sind in tonigen Formationen bedeutende Auswirkungen bei der Testplanung und bei der Interpretation der Testresultate die Folge. Offensichtlich kann die Grösse der hydraulischen Druckhöhe empfindlich von den chemischen Verhältnissen der Bohrlochflüssigkeit abhängen. Dies stellt die übliche Praxis, jeweils Süsswasser als Testlösung zu verwenden, in Frage. Nichtpolare und nichtreaktive Lösungen wären wohl eher geeignet. Umgekehrt könnten die chemischen Eigenschaften der Testlösung als Variable bei der Entwicklung verschiedener Testmethoden in Tongesteinen in Betracht gezogen werden.

Im Verlauf dieser Studie wurde zwischen der chemisch-osmotisch induzierten Wasserbewegung (gekoppelt) und den physico-chemischen interpartikularen Quellvorgängen ein interessanter Zusammenhang aufgedeckt. In der unmittelbaren

Umgebung des Bohrlochs sind die beiden Mechanismen praktisch nicht unterscheidbar, da in beiden Fällen Wasser der Testzone in die umgebende Formation gezogen wird und beide Prozesse empfindlich auf die chemische Zusammensetzung des Testwassers reagieren. Dies wiederum bedeutet, dass beide Prozesse durch eine einzige, umfassende Theorie beschrieben werden könnten. Dabei dürfte es kein Zufall sein, dass für Tone sowohl die Wirkung als osmotische Membrane, als auch deren hohe Quellfähigkeit, durch dieselben Eigenschaften verursacht werden.

Verschiedene Themenkreise, die in diesem Bericht angeschnitten wurden, sind von derart ausschlaggebender Auswirkung für jede hydrogeologische Charakterisierung eines Tongesteins,dass sich einerseits weitere Feld-und Laboruntersuchungen geradezu aufdrängen und anderseits der theoretische Rahmen für das Verständnis der Vorgänge weiterentwickelt werden sollte. Zur weiteren Abkläung werden drei Stossrichtungen empfohlen: (a) Entwicklung einer speziellen Testmethodik für Bohrungen in Tongesteinen und Erstellen der dazu notwendigen theoretischen Ansätze für die Auswertung: (b) Entwicklung eines relativ einfachen, möglicherweise eindimensionalen, numerischen Modells um eine Sensitivitätsanalyse der transienten Strömung aufgrund der hydraulischen und chemischen Gradienten durchführen zu können: und (c) weitere Abklärungen über tektonische Bewegungen, die damit verbundenen Deformationen des Opalinus-Tons und mögliche Auswirkungen auf die regionale Hydrogeologie. Für spezifische Hinweise sei auf Kapitel 9 verwiesen.

SUMMARY

At a meeting of NAGRA and BGS representatives, held at Keyworth on May 24th, 1989, a number of processes were examined which might, conceivably, be responsible for the anomalously low hydraulic heads observed in borehole testing in the Opalinus Clay. An anomalous hydraulic head was defined as a head which cannot be interpreted in terms of normal gravitational advective flow under steady-state conditions.

In examining the problem it was apparent that three types of process might be invoked, the first associated with long-term transient flow, the second with coupled flow (in particular osmotically-driven flow), and the third with borehole effects such as swelling, the perturbation of *in situ* stresses and plastic deformation of the borehole walls.

This report represents a preliminary study of these identified processes. The approach taken has been to examine each type of process individually, first at a general level, and then with specific reference to the Opalinus Clay. Simple theoretical frameworks are established and a number of scoping calculations are performed in an attempt to distinguish first- and second-order effects. Baseline hydrogeological, geotechnical and geochemical parameters for the clay-shale are obtained from the available test data or are calculated from the known interelationships between properties. Where no data exists, "generic shale" properties are taken from the literature.

What emerges from this study is that, although the mechanisms of groundwater movement in mudrocks are at present uncertain, certain processes could have such profound hydrogeological effects that their significance goes way beyond the interpretation of borehole test results, with repercussions in the development of a strategy for site characterisation, in modelling and in safety assessment.

Our calculations suggest that long-term transient flow may be occurring in the Opalinus Clay as a consequence of the stress changes associated with the process of overconsolidation and the removal of sedimentary cover by erosion. In addition, based on the strong evidence for neotectonic deformation of the crust in many areas of Switzerland, anomalous hydrogeological conditions may develop in low permeability rocks such as Opalinus Clay as a result of stress- or strain-induced pore pressure changes.

The general picture which emerges from our calculations on the overconsolidation mechanism is one in which fracture flow fairly rapidly re-equilibriates after exhumation but re-adjustments of the water content of the intact mudrock may occur over a much longer time period, constrained somewhat by diagenetic bonding of clay minerals and the presence of cements. Thus fracture flow might be regarded as being essentially out of phase with flow in the intact clay (matrix flow).

The low porosity and generally high clay content of Opalinus Clay suggest that it might also act as an efficient semi-permeable membrane supporting osmotically-driven flow not only across the stratum but also within it. Our calculations show that osmosis can have large effects on hydraulic head measurements and these must be considered when testing in mudrocks. Coupled flow may also be extremely important in modelling groundwater movement in mudrock environments. If we acknowledge osmotic flow to

be a significant component of total flow in mudrocks, then long-term transient behaviour must be viewed as a response to *hydraulic and chemical disequilibria.*

Deep burial and subsequent exhumation of the Opalinus Clay have endowed it with a marked "thirst" for water. This propensity to draw in water can be viewed in simple mechanical terms as a consequence of rebound from overconsolidation or in chemical terms as a suction associated with the processes of osmosis and/or hydration operating at the microscopic scale. Large suctions (negative pore pressures) may develop in near surface material. The presence of these suctions is entirely consistent with the observed high swelling capacity of the clay. It is also fundamental to our discussions on long-term transients and osmotically-driven (coupled) flow within mudrocks.

Thus we see that in mudrocks, particularly highly compacted clay-shales like the Opalinus Clay, there are a considerable number of reasons why the hydraulic head (determined in the usual way) might not be interpretable in terms of gravitational flow under steady state conditions. In fact, it is difficult to argue the case that the head should *not* be "anomalous" in many mudrock environments.

Borehole effects associated with the mechanical, thermal and chemical changes occurring in the formation during drilling and testing can have a significant influence on the measured hydraulic response during testing. Although swelling due to the introduction of fresh water is probably the most significant effect, in deep boreholes perturbation of the *in situ* stress field and plastic deformation of the rock may have a considerable effect on the pore pressure distribution around the borehole. If the measuring system has low compliance, the measurements may also be affected by time-dependent borehole closure.

When we examine the particular case of the anomalous hydraulic heads in borehole RB26B at the site of the proposed Wisenberg Tunnel in the Homburger Tal (see Section 6.2), then we cannot totally exclude very simple explanations for the response such as the presence of air in the test zones. However, these anomalous heads can also be explained (semi-quantitatively) by assuming that the total head at each point in this borehole is given by:

$$\mathbf{h} = z + h_{pp} + h_{sp}$$

where z is the elevation head, h_{pp} is the pressure head in the macropores (associated with hydrostatic pore pressure), and h_{sp} is the solute head in the macropores. If this scenario is correct, then it has very important implications to the design and interpretation of hydraulic tests in mudrock formations. The most obvious consequence is that the *magnitude of the hydraulic head determined in such tests is likely to be sensitive to the solute chemistry of the test fluid*. This calls into question the common practice of using *fresh water* as the test fluid. Non-polar, non-reactive liquids might be more suitable for this purpose. Alternatively, the chemistry of the test fluid might be regarded as a *variable* in the development of a testing methodology for mudrocks.

An interesting relationship emerges in this study between chemico-osmotic (coupled) flow in a mudrock and the physico-chemical process of interparticle swelling. In the vicinity of a borehole, the two processes are virtually inseparable. Both provide a mechanism by which water in the test zone is drawn into the formation and both are sensitive to the chemistry of the test fluid. This suggests that both processes might be describable in terms of a single unified theory. It is probably no coincidence that the characteristics of a clay which render it an efficient osmotic membrane are identical to those which endow it with a high capacity to swell.

The issues raised in this report are so important to hydrogeological site characterisation in mudrock environments that additional field and laboratory studies are clearly demanded, backed up by a parallel development of the theoretical framework. We distinguish three priority areas: (a) Development of a borehole testing methodology specific to mudrocks, together with the necessary theoretical models for reduction of the test data; (b) Development of a relatively simple, possibly one-dimensional, numerical modelling capability to allow sensitivity analyses to be performed on transient flow under hydraulic and chemical gradients; and (c) Further examination of the implications of tectonic deformation to the regional hydrogeology of the Opalinus Clay. Specific recommendations are made in Chapter 9.

KEYWORDS:

HYDROGEOLOGY, CLAY, CLAY-SHALE, HYDRAULIC TESTS, ANOMALOUS HEADS, TRANSIENT FLOW, UNDERPRESSURE, OVERCONSOLIDATION, OSMOSIS, SWELLING

LIST OF CONTENTS

LIST OF FIGURES

Fig. 8 Coupled and direct flow phenomena relating to the movement of fluid, heat, electrical current and ions. Direct flow mechanisms are shown in bold typeface and appear on the main diagonal. The associated physical law is given in italics. Coupled mechanisms lie off the main diagonal (adapted from MITCHELL, 1976).

Fig. 9 Hydrogeological completion of Borehole RB 26B at the site of the proposed Wisenberg Tunnel, Homburger Tal.

Fig. 10 Head measurements in Borehole RB 26B, site of the proposed Wisenberg Tunnel: Data for 1988. Total head **h** is depicted, with datum at ground surface (440.90 metres above mean sea level).

Fig. 11 Head measurements in Borehole RB 26B, site of the proposed Wisenberg Tunnel: Data for 1989. Total head **h** is depicted, with datum at ground surface (440.90 metres above mean sea level).

Fig. 12 Diagram illustrating local variation of the pressure head and solute head components of total head in a mudrock and the relationship between measured pressure head h_{pmeas} and the *in situ* parameters (adapted from MITCHELL, 1976).

Fig. 13 Idealised situation of a clay membrane separating fresh water and saline aquifers. Zero vertical groundwater flux is assumed. The measured pressure heads h_{pmeas} in this system would suggest that a total head gradient exists at the clay / saline aquifer interface. In the absence of information on solute chemistry we would incorrectly assume that a vertical (upward) flux exists across this interface.

Fig. 14 A simple one-dimensional analogue for the hydro-chemico-mechanical responses of a swelling mudrock. The total stress σ on the mudrock is borne by the interparticle stress σ_i', the pore water pressure u_p, and the net physico-chemical forces per unit area acting between the clay platelets (R_{diff} - A). Pressure u_0 measured in a hydrogeological test is influenced by osmosis.

LIST OF TABLES

1 INTRODUCTION

NAGRA is currently investigating the feasibility of constructing an underground repository in Switzerland for the disposal of radioactive wastes. Two geological settings are under consideration, the crystalline basement of northern Switzerland with granite or gneiss as possible host-rocks, and a sedimentary rock environment with the Opalinus Clay (Jurassic) and Lower Freshwater Molasse (Tertiary) as the priority formations for investigation.

Characteristics which suggest the Opalinus Clay to be a suitable host-rock for the disposal of radioactive waste include the low permeability (due to high clay content), the largely uniform lithology of the strata and its predictability over large areas, good swelling capacity which provides the potential for the sealing of fractures, and desirable sorption properties (NAGRA, 1988).

As an essential part of the safety assessment, NAGRA is seeking to quantify groundwater flow within the Opalinus Clay and adjacent formations. The lithological and structural characteristics of the clay have been defined and mathematical models of the regional hydrogeology have been developed assuming steady-state Darcy flow. A programme of hydrogeological testing in boreholes, geochemical and hydrochemical studies and physical property measurements on cores is being undertaken.

Four very important issues must be addressed in modelling groundwater movements in mudrock environments:

- Is advection within a mudrock largely confined to the fracture network or does water movement in the intact rock (matrix flow) constitute a significant proportion of the total flow?

- Can we assume, as we do in more permeable media, that the hydrogeological system can be modelled in terms of steady-state flow or must we acknowledge the existance of long-term transient flow in these low permeability rocks and develop the models accordingly?

- Is groundwater flow within a mudrock driven exclusively by hydraulic gradient or must we consider chemical and possibly thermal gradients and the phenomenon of coupled flow?

- When considering flow in a mixed sedimentary sequence, does a mudrock stratum behave as a semi-permeable membrane separating more permeable strata and supporting coupled flow across it?

Unfortunately the questions raised do not have simple answers. Our current understanding of groundwater movements in mudrocks is based, in the main, not on direct measurements, which are exceedingly difficult in such low permeability media, but on inferences drawn from hydrogeological, geochemical and geotechnical evidence. Certain questions can never be answered by field measurements alone. For example, in view of the time-scale involved, it is quite impossible to demonstrate whether present conditions represent steady-state flow in equilibrium with stable boundary conditions or whether they are slowly transient in response to natural geological changes. The best that we can do is to form a reasoned judgement on this matter based on an assessment of the available evidence. It is also seldom possible to establish definite cause and effect in complex geological systems.

Thus, the main difficulty in demonstrating that coupled flow is operating at a locality is that the field evidence can often be explained by invoking other equally probable mechanisms.

The issues raised above have considerable repercussions not only in regional hydrogeological modelling but also in the development of a strategy for site characterisation and in the design and interpretation of hydrogeological tests.

The logistical difficulties of hydrogeological testing in low permeability media are well known and include the special demands placed on instrumentation, in terms of measurement resolution, and the very prolonged duration of the flow transients. However the measured responses are regarded as being exactly analogous to those in more permeable media, and the same interpretation methods are employed. Three assumptions are implicit in current hydrogeological testing practice. The first is that flow is driven solely by hydraulic gradient and total head is divisible into pressure and elevation head components, the second is that borehole effects (other than skin-effects

and quantified temperature and pressure history effects) are small and can be ignored, and the third is that the test fluid is non-reactive with the formation. Based on our current understanding of the geotechnical behaviour of clays and shales, these assumptions appear highly suspect. We ask the following questions, which are fundamental to hydrogeological testing in mudrocks:

- If we acknowledge that coupled (non-hydraulic) flow is occurring in a mudrock, then what quantity is actually being measured when we attempt to determine hydraulic head in a down-hole hydrogeological test?

- What precisely do we mean by the terms pressure head and pore pressure in a mudrock?

- Do borehole effects associated with the mechanical, thermal and chemical changes occurring in the formation during drilling and testing have a significant influence on the measured hydraulic response? Is borehole closure likely to be a major factor?

- If we introduce fresh water into a mudrock during hydrogeological testing, then it is highly probable that the physico-chemical process of swelling will occur in the material adjacent to the borehole. What effect will this process have on the hydrogeological measurements? Can we regard swelling as just another problem in testing or is the process fundamental in determining water movements in clays and shales? Are swelling, rebound and coupled flow inextricably linked?

In this report we briefly examine each of these issues with specific reference to the Opalinus Clay. Present knowledge of the mechanisms of groundwater flow in mudrocks is very incomplete and is dispersed throughout the geoscience and engineering literature. Specific contributions have been made in soil and rock mechanics, petroleum geology, hydrogeology, and clay mineralogy, although the approaches of these various disciplines have yet to be fully integrated. The theoretical framework for the analysis of problems is based largely on the concept of the simple deformable porous medium, and since some important characteristics of clays (including clay-water interaction) are not considered, models based on currently available theory

are probably inadequate to decribe the long-term hydrogeological behaviour of compacted clays and shales.

2 OPALINUS CLAY - GEOLOGY AND PROPERTIES

The Opalinus Clay (named after the ammonite Leioceras opalinum) represents the lowest stratigraphical unit of the Dogger (Middle Jurassic) and is found extensively in the entire Tabular and Folded Jura of Switzerland. It consists of a relatively monotonous sequence of grey to grey-black mica-rich marine clay-shales with intercalated beds and lenses of sands and silts and calcareous horizons. The thickness varies from 80 to 120 m. It is generally poorly exposed at the surface except in clay pits where it is excavated for industrial purposes.

Fig. 1 shows potential areas for future investigation within the Opalinus Clay identified by NAGRA. Fig. 2 shows a general geological cross-section (= A in Fig. 1) through central and northern Switzerland (THURY and DIEBOLD, 1987). Fig. 3 gives a geological cross-section (= B in Fig. 1) showing the Permocarboniferous troughs, Tabular and Folded Jura and the edge of the Molasse Basin (O = Opalinus Clay) (DIEBOLD, 1986).

The potential region for a shallow repository, around 300 - 600 m deep, extends from Schaffhausen to Rafzerfield (= R in Fig. 1) in the southern part of the Aargau Jura. Regions which would be suitable for a repository at a depth in the range 600 - 1200 m are the Zurich Weinland (= ZW) and an area just north of Laegeren (= NL).

NAGRA have drilled three deep boreholes intersecting the Opalinus Clay, at Schafisheim (= SHA in Fig. 1), Riniken (= RIN) and Weiach (= WEI) and, based on detailed logging of the cores, the Opalinus Clay has been sub-divided into six lithological units (BLAESI, 1987).

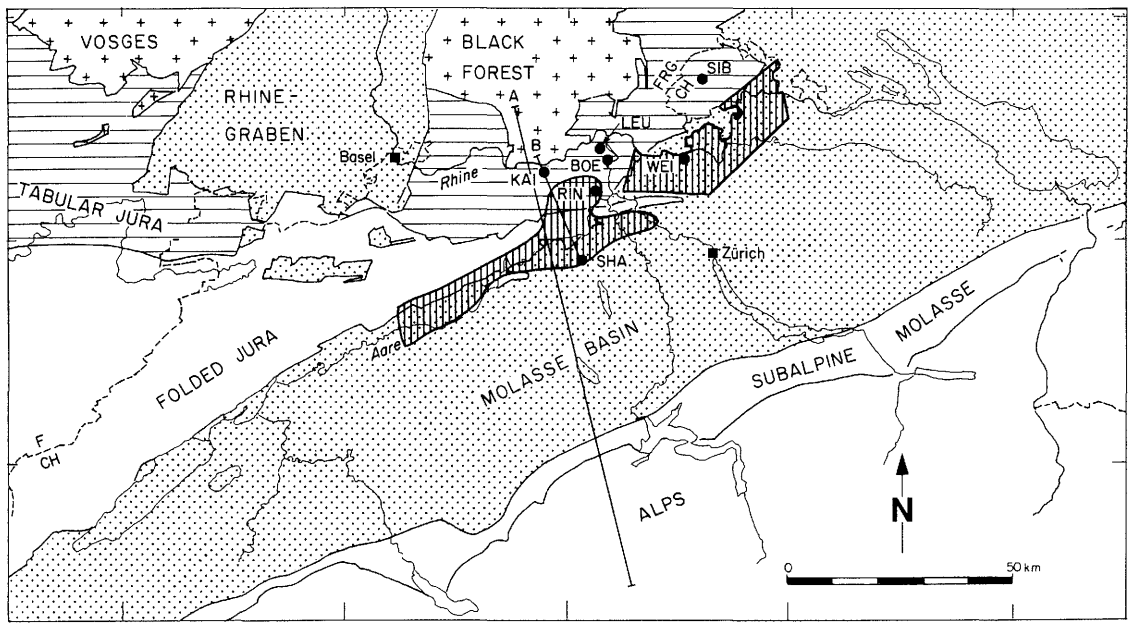


Fig. 1: Simplified geological map of central and northern Switzerland showing potential future investigation areas within the Opalinus Clay; see text for abbreviations. A: Cross-section Fig. 2; B: Cross-section Fig. 3.

In addition, NAGRA is collecting and analysing information from other ongoing projects in Switzerland which can be considered as analogues to improve basic knowledge about the Opalinus Clay. One of these projects is a planned railway tunnel (Wisenberg Tunnel) between Sissach and Olten. During the Wisenberg Tunnel Project several boreholes were drilled by the Swiss Federal Railway Company to obtain geological and hydrogeological information. One borehole (RB26B in Fig. 1) intersects the Opalinus Clay in the Homburger Tal.

The maximum depth of burial of the Opalinus Clay in the Homburger Tal is estimated to be 400 m. Valley erosion may have removed up to 350 m of the overlying sediments suggesting that the clay is in this area is heavily overconsolidated. In the Zurich Weinland the clay has been buried several hundred metres deeper than the Homburger Tal area and is probably only lightly overconsolidated (NAGRA, pers. comm., 1989).

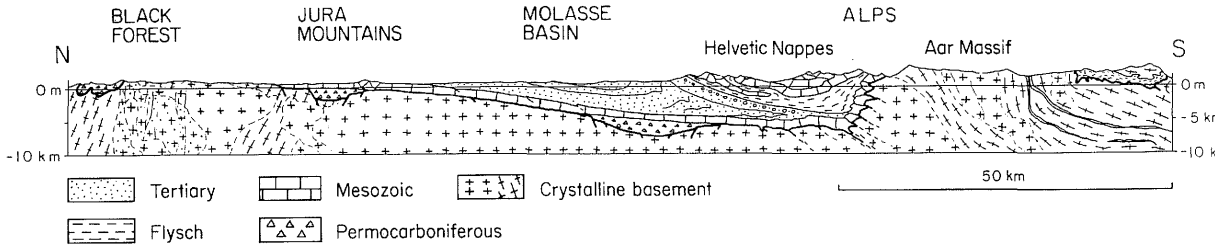


Fig. 2: Cross-section A through central and northern Switzerland (THURY and DIEBOLD, 1987).

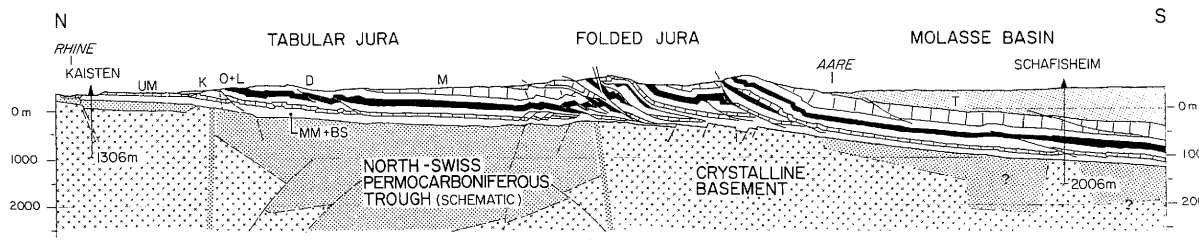


Fig. 3: Cross-section B showing Permocarboniferous troughs, Tabular and Folded Jura, and edge of Molasse Basin; T = Tertiary, M = Malm, D = Dogger, O+L = Opalinus Clay and Liassic, K = Keuper, UM = Upper Muschelkalk, MM-BS = Middle Muschelkalk to Bunter Sandstone (DIEBOLD, 1986).

In the Riniken and Schafisheim boreholes, the lower part of the Opalinus Clay is subject to high horizontal stresses of tectonic origin. Fractures observed in core often exhibit polished or striated surfaces (slickensides) indicating that the clay may have functioned as a shearing horizon during tectonic movement. Soft clay infillings have been noted in some fractures in core from the Riniken borehole.

Fig. 4 shows a schematic stratigraphical and hydrogeological profile for northern Switzerland (GAUTSCHI, 1989). The groundwater flow regime is complex and has

been characterized by several hydrodynamic models (KIMMEIER et al., 1985; NAGRA, 1988). The main recharge areas for the aquifers are situated in the Alps, the Folded Jura and in the Black Forest. The main discharge occurs to the Rhine River along the Swiss / German border and to the Aare River south of the Folded Jura. In the Molasse Basin (Fig. 1), infiltration and exfiltration are strongly influenced by local flow systems and therefore display a rather complex pattern (GAUTSCHI, 1989; NAGRA, 1988).

When exposed at the surface, the Opalinus Clay exhibits a conspicuous orthogonal fracture system with well-developed joints, perpendicular to the bedding, at typically 0.5 to 1 m spacing. The joint surfaces and bedding planes are often covered with an ochre-brown limonite coating which is indicative of the movement of oxygen-rich meterioric water within these discontinuities.

No iron oxide coatings have been observed in deep borehole material. There is a possibility that sub-vertical water flow occurs along steep disturbed zones and vertical fractures and sub-horizontal flow along bedding joints or gliding joint systems. Hydrogeological tests in boreholes suggest hydraulic conductivity (horiz.) to be in the range 10^{-12} to 10^{-15} $m.s^{-1}$, although no exact value can be given because of non-ideality of the pressure responses.

Due to the low permeability of the clay no field water samples have been obtained. Leaching tests on deep borehole material indicate porewaters with 0.2 $mol.l^{-1}$ Cl^- and squeezing experiments on material from the Mt. Terri Tunnel yield 0.28 $mol.l^{-1}$ Cl^-. NaCl waters with 1.28 $mol.l^{-1}$ Cl^- have been sampled from the underlying Liassic / Keuper formations at a depth of 1400 m in oil wells. Flushing by fresh water may have occurred in near surface Opalinus Clay in the Folded Jura.

Mineralogically the Opalinus Clay comprises quartz (11-23%), calcite (3-67%), siderite (0-40%), ankerite (0-6%), feldspar (0-5%) and clay plus accessory minerals (25-75%). Accessory minerals include pyrite (1-2%) and organic carbon (0-2.7%). Clay mineralogy comprises illite (40-50%), kaolinite (15-45%), chlorite (5-25%), and mixed layer illite-smectite (5-20%) (NAGRA, 1988).

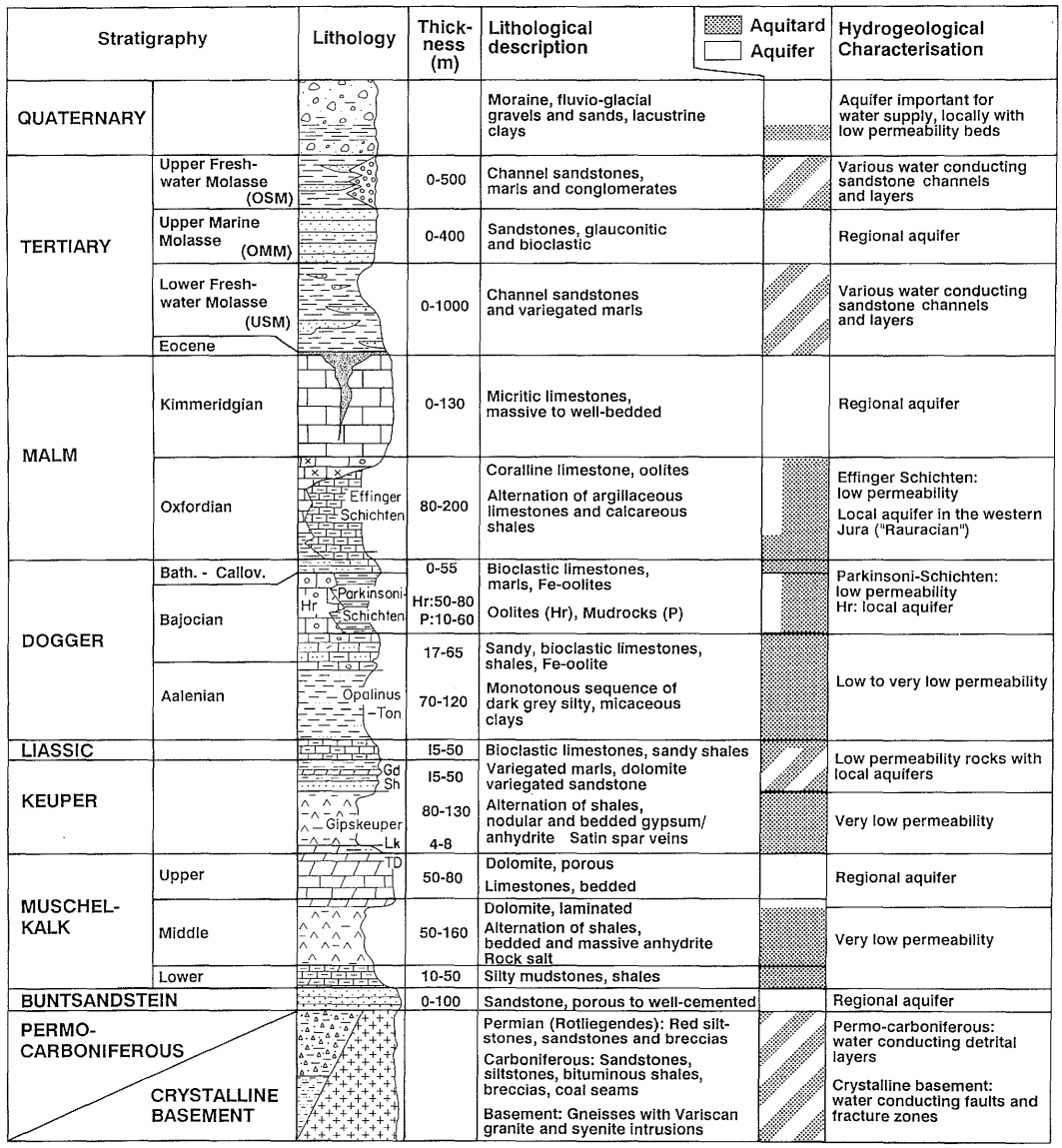


Fig. 4: Schematic stratigraphical and hydrogeological profile for northern Switzerland; Hr = Hauptrogestein, Gd = Gansinger Dolomite, Sh = Schilfsandstein (sandstone), Lk = Lettenkohle, TD = Trigonodus Dolomite (GAUTSCHI, 1989).

The specific surface of the clay fraction is typically 140 $m^2.g^{-1}$ and the cation exchange capacity (CEC) typically 25 $meq.100g^{-1}$.

The geotechnical properties of the clay are strongly dependent on water content, which in turn, is dependent on effective stress. High pressure triaxial experiments on natural

and artificially recompacted clay are reported by NUESCH (1988). Destressed material demonstrates a high swelling capacity with swelling pressures in the range 0.7 to 2.2 MPa (MADSEN and MULLER-VONMOOS, 1985). Very limited data exist for the strength parameters of the Opalinus Clay. Uniaxial compressive strength in the range 8 to 36 MPa is believed representative. Plastic limit is in the range 17-26%, liquid limit 40-80% and plasticity index 23-54% (BUCHER, 1975).

3. BASIC PARAMETERS

3.1 Porosity

Porosity is a basic parameter in hydrogeology describing the open, fluid-filled voids in a rock. Numerically total porosity is the ratio of the volume of voids to the total volume and, recognizing that the value we assign to a given rock will depend on scale at which we examine it, we select a representative elementary volume (REV), appropriate to the problem under examination, for our definition. Furthermore, we recognize that not all the pore spaces are interconnected and capable of conducting fluids and, conventionally we define an additional parameter, the effective porosity, as that component of total porosity that is capable of participating in the advection process.

In highly-compacted clays and shales we encounter great difficulties in both the definition and the measurement of total porosity and these difficulties become even more acute when we attempt to distinguish effective porosity. Our problems stem from the microscopic scale of the voids in compacted clays and the complex clay-water interactions near the clay mineral surfaces. It is possible to distinguish four types of water in clays, intermicellar water within the sheet structure of certain clay minerals, notably smectites, highly adsorbed water present as thin molecular films around the clay minerals, loosely adsorbed water associated with the clays but more remote from the mineral surfaces, and finally "free water" which is sufficiently remote from the clay particles to be unaffected by them. Only loosely adsorbed and free water can participate in the advection process.

If we assume that water is present, in the main, as films around predominantly platy clay minerals, we can estimate the average half distance d (nm) between these clay platelets in

a saturated clay using the following relationship, which is slightly modified from the expression given in YONG and WARKENTIN (1975)

$$d \approx \frac{m}{\gamma_w \cdot C \cdot S} \cdot 10^3 \quad \text{(nm)} \qquad (1)$$

where γ_w is the density of water ($Mg.m^{-3}$), m is the water content determined at the normal 105°C drying temperature and expressed as the ratio of weight of water to dry weight of solids (%), C is the clay fraction expessed as the dry weight of clay to the dry weight of solids (%) and S is the specific surface of the clay fraction ($m^2.g^{-1}$).

Performing this calculation for the Opalinus Clay, taking m in the range 3.2 - 5.2% (see Section 4.2) and $\gamma_w = 1\ Mg.m^{-3}$, $S = 140\ m^2.g^{-1}$ and C = 65%, we find

$$d \approx 0.35 - 0.57 \quad \text{nm}$$

which suggests that the clay platelets are separated by an "average distance" which is in the approximate range 0.70 - 1.14 nm. If we assume that some of the water resides in macropores, then this separation will be even smaller. *In view of the fact that the van der Waals molecular radius of water is 0.28 nm, the difficulty of defining an effective (or interconnected) porosity for the advection process, other than fracture porosity, becomes apparent.*

We define microporosity, for our purposes, as the space occupied by intermicellar and adsorbed water, and macroporosity as the space occupied by free water. Then total porosity is then simply the sum of the microporosity and the macroporosity.

We note that in the basic relationship (1), above, we assume that the density of highly adsorbed water is equal to that of free water and that a standard moisture content test removes all free and adsorbed water from the clay. The general picture does not change significantly if we were to adopt alternative assumptions for our analysis.

3.2 Potentials, Heads and Pore Pressures

HUBBERT (1940) defines potential as "a physical quantity, capable of measurement, whose properties are such that flow always occurs from regions in which the quantity has higher values to those in which it has lower, regardless of the direction in space". For advective flow in a porous medium, the quantity which satisfies this definition is the mechanical energy per unit mass of water referred to an arbitrary reference state.

If we take z to be a vertical coordinate with the positive direction upwards and take as our reference state water at elevation z = 0, at rest, and at pressure u_0, then the work done per unit mass in raising the water isothermally to elevation z and pressure u, and accelerating it to velocity v is

$$\phi = g z + \frac{v^2}{2} + \int_{u_0}^{u} \frac{du}{\gamma_w(u)} \qquad (2)$$

where γ_w is the density of water. Since the water velocity is usually extremely small in a porous medium, we normally drop the second term in (2). If we assume that the water is incompressible, then (2) simpifies to

$$\phi = g z + \frac{u - u_0}{\gamma_w} \qquad (3)$$

or

$$\phi = \phi_z + \phi_p \qquad (4)$$

where ϕ_z is the elevation (gravitational) potential and ϕ_p is the pressure (matrix) potential. Since the acceleration due to gravity g varies little from place to place over the surface of the earth, we can divide (3) by g and still meet Hubbert's requirement for a potential. Using gauge pressures (u_0 = atmospheric = 0) we obtain

$$\frac{\phi}{g} = z + \frac{u}{g\,\gamma_w} \tag{5}$$

or

$$\mathbf{h} = z + h_p \tag{6}$$

where **h** is the total head, z is the elevation head and h_p is the pressure head.

Chemico-osmotic flow is widely recognized in clay soils and a component potential is often included in the definition of total potential (YONG and WARKENTIN, 1975), thus

$$\phi = \phi_z + \phi_p + \phi_s \tag{7}$$

where ϕ_s is often termed the solute potential, defined as the work done per unit mass of water in transporting reversibly and isothermally an infinitesimal quantity of water from a pool of pure water, at a specified elevation and at atmospheric pressure, to a pool containing a solution identical in composition to the pore water, at the same elevation and pressure as the reference pool.

By analogy with (6)

$$\mathbf{h} = z + h_p + h_s \tag{8}$$

We note that ϕ_s and h_s are negative quantities since the flow direction is towards the more concentrated solution. We will explore the implications of the solute head component of total head in some detail in Chapters 6 and 7.

4 LONG-TERM TRANSIENT FLOW

It is a common assumption in hydrogeology that regional groundwater movements occur under steady-state conditions. The rationale behind this assumption is that the perturbations in stress, strain and temperature associated with burial, folding, faulting,

erosion and a multitude of additional geological processes occurred at a time sufficiently remote from the present day that their resulting flow transients have long since died out. Furthermore, ongoing geological processes are considered to proceed at such slow rates that they cannot produce significant departures from steady-state.

However, there is a growing volume of evidence to suggest that, in low permeability formations, flow transients resulting from past geological activity may be of sufficient duration and magnitude to dramatically influence the current groundwater flow pattern (NEUZIL, 1986). *This is particularly true of clay and shale formations which have the attributes of exceedingly low permeability combined with relatively large compressibility.*

Overpressuring in oilfield shales has often been ascribed to long-term transient responses resulting from changes in stress, strain or temperature within the shales, or to diagenetic processes such as clay transformation and cementation. One of the earliest and perhaps the simplest of explanations for overpressure is that the rate of deposition of the column of sediments overlying the clay stratum was so high that pore pressures developed during compaction were unable to dissipate. This theory has been termed "*compaction disequilibrium*" and can be simply interpreted in terms of the rate of change of total stress acting on the clay during burial, the rate of change of pore pressure, and the rate of drainage by advective flow (RUBEY and HUBBERT, 1959; BREDEHOEFT and HANSHAW, 1968; SMITH, 1971; KOJIMA et al., 1977). In the "*aquathermal theory*" of overpressure, increases in temperature occasioned by increased depth of burial or other processes are invoked as the source of abnormal pore pressures and the resulting long-term flow transients (LEWIS and ROSE, 1970; BARKER, 1972; MAGARA, 1974; 1975). Similar effects to those envisaged in the "compaction disequilibrium" theory could be produced by the high stress- and strain-rates imposed on a shale during a tectonic episode (BERRY, 1973). Clay mineral transformation, in particular the loss of intermicellar water by smectites, during burial has also been suggested as a mechanism of overpressuring. A basic assumption of this theory is that highly adsorbed water has a significantly larger density than that of free water, and the release of such water from the clay will therefore result in a net volume increase (POWERS, 1967; BURST, 1969). COLTEN-BRADLEY (1987) indicates that the loss of intermicellar water by smectite does not occur at the temperatures and pressures associated with normal burial and that changes in the hydration state are primarily controlled by changes in pore fluid salinity or layer charge, casting doubt on

this mechanism of overpressuring. Furthermore, the basic assumption that adsorbed water is denser than free water is controversial (ANDERSSON and LOW, 1958; MARTIN, 1962; BORGESSON et al., 1988).

Almost every mechanism postulated as the cause of overpressure could, can by reverse arguments, be invoked as possible source of underpressure or anomalously low pore pressures. Thus we have underpressure resulting from destressing during erosion of the sediment column or the removal of ice loading (RUSSELL, 1972; DICKEY and COX, 1977; NEUZIL and POLLOCK, 1983; TOTH and MILLAR, 1983), from temperature decreases, and from dilatancy (the opposite of compaction) and fracturing associated with tectonic activity.

Thus we rapidly reach the conclusion that, *given an adequate thickness of low permeability clay, then any geological process that is capable of changing the stress state or temperature of the clay, or of deforming and fracturing it in any way, or of altering the physico-chemical state of the adsorbed water layers, could also be invoked as the source of a long-term transient hydrogeological response.* It also seems probable that, since numerous geological processes can operate simultaneously, long-term transients associated with each process can become superimposed.

Whether or not a particular geological process can be offered as a reasonable explanation of an abnormal pressure situation will depend primarily on the *predicted time duration* of the hydrogeological transient produced by that process, and on the *predicted magnitude* of the effect at the present day. Thus, in order to advance any further with this topic, we are forced into calculations of transient flow reponse over very protracted time periods with all their attendant difficulties.

4.1 Transient Flow in a Deforming Mudrock

The traditional starting point in the development of the governing equations for transient flow in a saturated porous medium is an examination of the conservation of water mass and solid mass in the deforming medium (BIOT, 1941; BEAR, 1972). The medium is envisioned as comprising a matrix of solid particles enclosing water-filled pores which, for the purposes of the analysis, are assumed to be interconnected. Thus, even at this most general level, we run into difficulties in applying available theory to compacted clays since the *existence of the highly significant adsorbed water layers is not*

acknowledged, and the interconnection of the macroporosity is by no means proven in compacted material.

Acknowledging that these deficiencies exist, we proceed to quantify density changes in the solid and water phases in terms of temperature, pressure and stress changes and the appropriate compressibility and thermal expansion constants. Implicit in these calculations is the assumption that the *mineralogical composition of the clay remains fixed over possibly prolonged time spans.*

At this point we introduce a constitutive equation describing the volumetric strain in terms of temperature, pressure and stress changes.

Studies on the compaction of shales reveal a general pattern with increasing depth of burial

$$e = e_1 - b_1 \log_{10}(z) \tag{9}$$

where e is the voids ratio, e_1 and b_1 are constants and z is the depth of burial (VON ENGELHARDT, 1960; RIEKE and CHILLINGARIAN, 1974). If we assume that the vertical effective stress σ_v' increases linearly with depth, then (9) can be re-written as

$$e = e_2 - b_2 \log_{10}(\sigma_v') \tag{10}$$

where

$$\sigma_v' = \sigma_v - u \tag{11}$$

and σ_v is the total vertical (lithostatic) stress due to the weight of the overburden and u is the pore pressure.

It is a common characteristic of soils and argillaceous rocks that the voids ratio / effective stress relationship during compaction differs considerably from that exhibited by these materials during unloading. In effect, only part of the porosity loss suffered

by these materials during compaction process can be recovered during unloading. A clay which has been compacted to an effective stress no higher than the current effective stress is called "*normally consolidated*" in soil mechanics terminology. This of course suggests that the clay has never been buried deeper than its present depth, nor has it been subjected to tectonic stresses which exceed present-day stresses. A clay

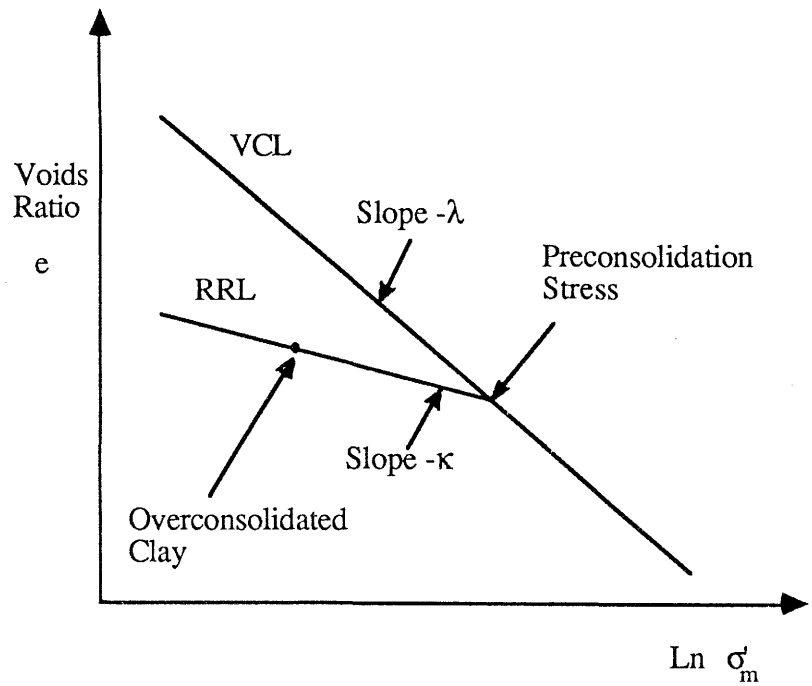


Fig. 5: Schematic plot of voids ratio against logarithm of mean normal effective stress for a mudrock undergoing burial and exhumation (VCL = Virgin Consolidation Line, RRL = Rebound / Reconsolidation Line). An "overconsolidated" mudrock is represented by a point on the RRL.

which has been compacted in the past to an effective stress greater than the current value is termed "*overconsolidated*" and the value of the maximum past effective stress is termed the "*preconsolidation stress*".

If we assume that no shear stresses are developed during burial and exhumation, then the voids ratio in the normally consolidated state may be described, in terms of *Critical State Theory* (SCHOFIELD and WROTH, 1968) by

$$e = e_a - \lambda \ln\left[\frac{\sigma_m'}{\sigma_{ma}'}\right] \tag{12}$$

and in the overconsolidated state by

$$e = e_b - \kappa \ln\left[\frac{\sigma_m'}{\sigma_{mb}'}\right] \tag{13}$$

where σ_m' is the "mean normal effective stress" (the mean of the three principal stresses minus the pore pressure), given by

$$\begin{aligned} \sigma_m' &= \sigma_m - u \\ &= \frac{1}{3}(\sigma_{11} + \sigma_{22} + \sigma_{33}) - u \end{aligned} \tag{14}$$

and $-\lambda$ and $-\kappa$ are the slopes of the "virgin consolidation line" (VCL) and "rebound / reconsolidation line" (RRL), respectively, in [e, ln(σ_m')] space, e_a is the voids ratio at effective stress σ_{ma}' on the VCL, and e_b is the voids ratio at σ_{mb}' on the RRL. A schematic of this idealised behaviour is given in Fig. 5.

With very great additional complication we could also examine shear stress effects on compaction and dilatancy using Critical State Theory. *However, the main points we wish to convey here are that the voids ratio and hence the porosity of an argillaceous rock are nonlinearly related to the effective stress and that the voids ratio / stress relationship varies from the normally consolidated to the overconsolidated states.*

The introduction of such nonlinearities into a general theory of transient flow in clays poses significant mathematical problems, although it is a necessary requirement if we wish to fully understand and accurately quantify long-term flow transients in argillaceous rocks. All efforts, to date, to model long-term transients in low-permeability media have linear elastic theory as their basis. *For this reason analytical solutions which have been obtained are valid for infinitesimal strains only, and because the problem has been "linearised", these solutions only hold for small*

changes in stress and temperature.

The mathematical framework for the analysis of transient flow in a saturated, linear elastic, porous medium is provided by Biot's theory of three-dimensional consolidation (BIOT, 1941; 1955).

Using Biot's theory as a basis, and assuming that the medium is *mechanically isotropic, relatively incompressible*, and that its volumetric strain can be decribed by the *linear thermoelastic constitutive equation*

$$\varepsilon = \frac{1}{K}\left[\sigma_m - \frac{K}{H} u\right] - \alpha_b (T - T_0) \tag{15}$$

the following relationships emerge (see Appendix A)

$$\frac{1}{R}\frac{\partial u}{\partial t} = \nabla \cdot (\underset{\approx}{\mathbf{K}}\, \nabla \cdot \phi^*) + \frac{1}{H}\frac{\partial \sigma_m}{\partial t} + \alpha_m \frac{\partial T}{\partial t} \tag{16}$$

$$\frac{1}{Q}\frac{\partial u}{\partial t} = \nabla \cdot (\underset{\approx}{\mathbf{K}}\, \nabla \cdot \phi^*) + \zeta \frac{\partial \varepsilon}{\partial t} + (\zeta \alpha_b + \alpha_m)\frac{\partial T}{\partial t} \tag{17}$$

where K is the drained bulk modulus, R, H, Q and ζ are Biot's poroelastic constants, α_b and α_m are thermal expansion coefficients, u is pore pressure, σ_m is the mean normal total stress, ε is the volumetric strain, and T is temperature. The term $(\nabla \cdot (\underset{\approx}{\mathbf{K}}\, \nabla \cdot \phi^*)$ is the volumetric flux of water written in terms of Darcy's law for compressible flow where $\underset{\approx}{\mathbf{K}}$ is the hydraulic conductivity tensor and ϕ^* is Hubbert's potential (Appendix A).

PALCIAUSKAS and DOMENICO (1989) present (16) and (17) as the basis of their discussions on pore pressures in deforming porous media and emphasise the difficulties in establishing values for Biot's constants which are representative of geological time-scales. SHI and WANG (1986) provide an additional perspective on pore pressure develoment in a deformable medium, including thermal effects, and give a clear account of the derivation of the governing differential equations. Useful additional references on poroelastic theory are CORAPCIOGLU and BEAR (1984) and

VERRUIJT (1984), the latter providing a soil mechanics perspective.

4.2 Estimation of Biot's Constants for the Opalinus Clay

Recognising the effective stress dependence of the linear poroelastic constants, we choose to calculate two discrete sets of "baseline" parameters, the first pertaining to shallow Opalinus Clay in the Homburger Tal region, and the second to the deeper and more highly stressed material in the Zurich Weinland. We shall refer to these sets simply as *"shallow parameters"* and *"deep parameters"*. In view of the uncertainties of the calculation procedure, and the lack of pertinent laboratory test results, we feel that additional sophistication cannot be justified at this stage.

From our current knowledge of the stress history of the Opalinus Clay in the Homburger Tal region it is apparent that the clay in this locality is heavily overconsolidated. In the Zurich Weinland the clay is present at much greater depth and is probably only lightly overconsolidated. We assume here that the relationship between voids ratio and effective stress for the clay at each locality is given by (13), but recognise that the preconsolidation stress in the Zurich Weinland may be significantly higher than in the Homburger Tal area. If this is indeed the case, then it suggests that the the current state of the clay at the two localities can be represented as two points each on a separate "rebound / reconsolidation line" (RRL) plotted in [e, $\ln(\sigma_m')$] space. Estimates of the current *in situ* stresses at the two localities also suggest unusually high horizontal stresses may be present, which are of course indicative of high shear stresses (NAGRA, pers. comm., 1988). However, in order to proceed without undue complication, we must ignore the effect of the shear stresses and assume that voids ratio is solely a function of mean normal effective stress.

WROTH (1972) shows that, assuming isotropy, the drained Young's modulus E for an over-consolidated clay can be estimated from the undrained modulus E_u using

$$E = \frac{2}{3} (1 + \nu) E_u \qquad (18)$$

where ν is the drained Poisson's ratio.

Site investigation tests for the proposed Wisenberg Tunnel (Homburger Tal region)

yield an average value for the undrained modulus E_u of 622 MPa with a range of 318 to 1158 MPa. The average moisture content and bulk density for this material are 5.3% and 2.49 $Mg.m^{-3}$, giving a voids ratio 0.144.

Tests conducted at the University of Karlsruhe on Opalinus Clay samples from a brickpit near Heiningen, Baden-Wuerttemberg give E_u values for intact materal of 300 MPa for σ_1 normal to bedding, and 1000 MPa for σ_1 parallel to bedding. The moduli were defined for strains of less than 1.5%, and the average water content and bulk density were 7.4% and 2.45 $Mg.m^3$, giving a voids ratio of 0.201 (WICHTER, 1979).

Since the two data sets are in reasonable agreement, we take the average value for Wisenberg Tunnel material of 622 MPa as the basis for our calculations. With the additional assumption that the drained Poisson's ratio ν is stress-independent and equal to 0.15 for a clay-shale (the mean and standard deviation of the 34 different shales tabulated in LAMA and VUTUKURI (1978) are 0.17 and 0.10, respectively), using (18) we estimate the drained Young's modulus E to be 477 MPa for this material.

The drained bulk modulus K is then simply estimated from E using the standard relationship

$$K = \frac{E}{3(1 - 2\nu)} \tag{19}$$

which gives K = 227 MPa for this material.

NUESCH (1988), in an experimental study on the strength and deformation of Opalinus Clay, examined the relationship between effective overburden stress and water content. When combined with the low pressure consolidation data of MADSEN (1976), Nuesch's test results indicate that

$$e = 0.2284 - 0.041 \ln\left[\frac{\sigma_m'}{\sigma_{mb}'}\right] \tag{20}$$

where σ_m' is the mean normal effective stress (MPa) and reference stress $\sigma_{mb}' = 1$ MPa. This provides a very approximate value for critical state constant κ of 0.041. If we assume that (20) relates to the same RRL as the Wisenberg Tunnel material, then for a voids ratio of 0.144 we obtain $\sigma_m' = 7.8$ MPa. Now, the drained bulk modulus K is related to the critical state constant κ by

$$K = \frac{1}{\beta} = \sigma_m' \left[\frac{1 + e}{\kappa} \right] \qquad (21)$$

where β is the bulk compressibility.

If we insert Wisenberg Tunnel values, K = 277 MPa, e = 0.144 and $\kappa = 0.041$, into (21) we obtain a second estimate for σ_m' of this material of 9.9 MPa which is tolerably close to the value determined above.

The analysis thus far indicates that shallow material from the tunnel site investigation may be subject to fairly high effective stresses or, alternatively, that the consolidation tests predict a κ value which is substantially larger than the field value. This question cannot be resolved without further experiments. The implications of high effective stresses at a shallow site are profound. Since the mean total stresses at the shallow depths of these site investigation boreholes are likely to be fairly low (<< 5 MPa), the suggestion is that the pore pressure must be negative. We interpret this as indicating that the Opalinus Clay at the Wisenberg Tunnel may be subject to large suctions. This is a difficult concept in classical hydrogeology. The implications are that *deep burial and subsequent exhumation of the clay have endowed it with a marked "thirst" for water.* As we shall see, this propensity to draw in water can be viewed in simple mechanical terms as rebound from overconsolidation or in chemical terms as a suction due to osmosis and / or hydration. The presence of large suctions is entirely consistent with the observed high swelling capacity of the Opalinus Clay. It is also fundamental to our discussions on long-term transients and coupled flow.

Since (20) and (21) predict somewhat different values for mean normal effective stress from the estimated constants, we must make minor adjustments to our baseline properties to produce a consistent set. By assuming an effective stress of 8.15 MPa and a voids ratio 0.142 (slightly less than the value quoted above), our estimates for K and κ become compatible with the other constants and we take these values to be

representative of the "shallow" clay.

If we assume that κ is constant for a clay in the overconsolidated state, then (21) will also hold the deeper material, even though the preconsolidation stress at other locations will probably differ from that of the shallow material. Lack of data limits any effort to construct a separate RRL for Opalinus Clay in the Zurich Weinland. *We therefore define our generic deeper material simply by its porosity which we assume to be 8%* (e = 0.087) and we assume that both (20) and (21) are applicable. The predicted mean normal effective stress for the deeper material is 31 MPa. The drained bulk modulus K, which is regarded as the fundamental parameter in subsequent calculations, is determined to be 834 MPa for the deeper material.

Biot's constant H can now be estimated for both parameter sets from

$$\frac{1}{H} = \frac{1}{K} - \frac{1}{K_s} = \beta - \beta_s \qquad (22)$$

where K_s is the bulk modulus of the solid particles. We assume an average compressibility β_s of 2.2 x 10^{-5} (MPa^{-1}) for the mineral constituents. The constant ζ is then easily determined from

$$\zeta = \frac{K}{H} = 1 - \frac{\beta_s}{\beta} \qquad (23)$$

Biot's constant R is given by

$$\frac{1}{R} = \beta_p + n\,\beta_w \qquad (24)$$

where the porosity n = e/(1+e). We assume the isothermal compressibility of water β_w to be 4.4 x 10^{-4} (MPa^{-1}). In Appendix A, the pore compressibility β_p is shown to be given by

$$\beta_p = \beta - \beta_s - n\,\beta_s' \qquad (25)$$

where β_s' is an independent grain compressibility parameter. BROWN and KORRINGA (1975) discuss the significance of this additional parameter and suggest that the overall bulk behaviour is insensitive to the numerical value we assign to it. In the absense of any information whatsoever on its value for the Opalinus Clay we assume $\beta_s' = 0$. Biot's constant Q can then be obtained from

$$\frac{1}{Q} = \frac{1}{R} - \frac{K}{H^2} \tag{26}$$

and the undrained bulk modulus K_u from

$$\frac{1}{K_u} = \frac{1}{K} - \frac{R}{H^2} \tag{27}$$

Finally, the constrained modulus of elasticity E_k is the reciprocal of the compressibility parameter m_v of one-dimensional consolidation theory

$$E_k = \frac{1}{m_v} = \frac{E(1 - \nu)}{(1 + \nu)(1 - 2\nu)} \tag{28}$$

Table 1 gives the calculated "best estimate" values for the "shallow" and "deep" parameter sets.

4.3 Estimation of Thermal Expansion Coefficients

With reference to Appendix A, the bulk cubical thermal expansion coefficient α_b of a porous medium can be related to the thermal expansion coefficents of the solids α_s and pore structure α_p by

$$\alpha_b = (1 - n)\,\alpha_s + n\,\alpha_p \tag{29}$$

Unfortunately the thermal expansion coefficients of the clay minerals are not well known. If we assume that $\alpha_s = \alpha_p$, then the bulk thermal expansion coefficient is equal to that of the solid particles ($\alpha_b = \alpha_s$) which is probably not unreasonable for a low

porosity clay. We take a generic linear coefficient of expansion for shale α_l of 2.2 x 10^{-6} $^oC^{-1}$ as the basis for our calculations. Assuming isotropy, α_b is equal to 3 x α_l and is approximately 7 x 10^{-6} $^oC^{-1}$.

Table 1: Baseline Poroelastic Parameters (Isotropic)

Symbol	Parameter	Opalinus Clay Shallow	Opalinus Clay Deep	Mudrock (Generic †)	Units
e	Voids ratio	0.142	0.087	-	-
n	Porosity ‡	12.5%	8.0%	-	-
m	Water content	5.2%	3.2%	-	-
κ	Critical state constant ‡	0.041	0.041	-	-
σ_m'	Mean Normal Eff. Stress	8.1	31	-	MPa
E	Drained Young's modulus	477	1752	-	MPa
ν	Drained Poisson's ratio ‡	0.15	0.15	-	-
E_u	Undrained Young's mod.‡	622	2285	-	MPa
K	Drained Bulk modulus	227	834	2128	MPa
K_u	Undrained Bulk modulus	13129	17796	10000	MPa
H	Biot's constant	228	850	2222	MPa
R	Biot's constant	225	825	1852	MPa
Q	Biot's constant	13031	17602	9091	MPa
ζ	Biot's constant	0.995	0.982	0.95	-
E_k	Constrained modulus	504	1850	-	MPa

† Values for generic mudrock taken from PALCIAUSKAS and DOMENICO (1989).
‡ These values are "fixed", all other values are calculated.

Table 2: Baseline Values for the α_m Expansion Coefficient for Opalinus Clay

Temperature (°C)	Best Estimate ($°C^{-1} \times 10^5$) Shallow	Deep
10	2.4	1.5
20	3.2	2.0
30	3.9	2.5
40	4.2	3.1
50	5.5	3.5

The α_m expansion coefficient of the transient flow equations is given by

$$\alpha_m = n\,\alpha_w - \alpha_b + (1 - n)\,\alpha_s \tag{30}$$

where α_w is the thermal expansion coefficient of water. Ignoring pressure effects we can evaluate α_w for a range of temperatures from

$$\alpha_w = \left[200 + 6.1\,(T - 10) \right] \times 10^{-6}\ °C^{-1} \tag{31}$$

where temperature T is expressed in °C. "Shallow" and "deep" parameter estimates for α_m are given in Table 2 for temperatures from 10°C to 50°C.

4.4 Pore Pressure Response during Undrained Loading

The simplest case to examine is that of isothermal, undrained loading which we interpret as a state of zero flux ($\nabla\cdot(\mathbf{K}\,\nabla\cdot\phi^*) = 0$) , in which case

$$\frac{\partial u}{\partial t} = \frac{R}{H} \frac{\partial \sigma_m}{\partial t} \tag{32}$$

Using our baseline values for the Opalinus Clay we find the ratio R/H to be 0.988 in the shallow case and 0.982 in the deep case. Since both ratios are very close to unity, we reach the important conclusion that *the entire incremental stress is supported by the pore pressure during undrained loading*. In this respect the response of the Opalinus Clay is more soil-like than rock-like.

4.5 Pore Pressure Response during Undrained Heating or Cooling

Assuming that the total stress remains unchanged during undrained heating, then the rate of change of pore pressure is described by

$$\frac{\partial u}{\partial t} = R \alpha_m \frac{\partial T}{\partial t} \tag{33}$$

As we have already shown, the expansion coefficient α_m varies considerably with temperature. For small temperature increments ΔT we may write

$$\Delta u = R \alpha_m \Delta T \tag{34}$$

where α_m is selected to be representative of the temperature range. At 20°C the pore pressure in our generic shallow Opalinus Clay is predicted to change by 0.007 MPa per °C change in temperature. The equivalent figure for deep material is 0.016 MPa per °C. At 40°C the predicted pressure changes are larger, 0.009 MPa per °C for the shallow material and 0.026 MPa per °C for the deep clay.

Our calculations assume that (a) the solid matrix is free to expand, (b) the mean normal total stress is not altered by the volume change of the matrix and, (c) the specific discharge relative to the *moving* matrix is zero. Although these "boundary conditions" are somewhat idealised, the predicted undrained response is likely to be close to that of a heated volume element within an extensive stratum of mudrock. If we were to assume that the solid matrix is not free to expand (i.e. constant mudrock volume during

heating), then the predicted pore pressure change would be much larger. The assumption of constant volume is, however, unrealistic for the field situation under examination.

4.6 Relative Significance of Loading/Unloading and Aquathermal Mechanisms

We assume that the aquathermal effect is primarily due to a temperature change ΔT produced by increasing or decreasing depth of burial Δz and that, as a first approximation,

$$\Delta T = \Delta z \cdot G_T \tag{35}$$

where G_T is the geothermal gradient. For constant stress, undrained conditions the pore pressure increment Δu becomes

$$\Delta u = R\, \alpha_m\, G_T\, \Delta z \tag{36}$$

The normal geothermal gradient is 0.02 to 0.03 $^{o}C.m^{-1}$. In northern Switzerland, G_T is expected to be somewhat greater (DIEBOLD and MULLER, 1985) and values in the range 0.041 - 0.046 were determined in the Weiach borehole (NAGRA, pers. comm., 1991). Taking 0.043 $^{o}C.m^{-1}$, we find that at 20°C the aquathermal effect produces a pore pressure change of 3.1×10^{-4} $MPa.m^{-1}$ for shallow Opalinus Clay and 7.1×10^{-4} $MPa.m^{-1}$ for deep clay.

If we assume the total stress gradient G_S to be 0.025 $MPa.m^{-1}$, then for isothermal, undrained conditions

$$\Delta u = \frac{R\, G_S}{H}\, \Delta z \tag{37}$$

and we find that the loading/unloading effect produces a pore pressure change of 2.5×10^{-2} $MPa.m^{-1}$ for both the shallow and the deep Opalinus Clay.

Thus, during burial or erosion, the pore pressure response to change in total stress (loading/unloading effect) is likely to be approximately 2 orders of magnitude greater than the response to change in temperature (aquathermal effect). We note that our simple undrained analysis is indicative of relative magnitude of these effects in the more complex transient flow problem. SHI and WANG (1986) modelled transient flow and the development of abnormal pressures by overloading and aquathermal effects using the finite element method, and their conclusions, which are based on data for a wide variety of rock-types, support our findings for the Opalinus Clay.

4.7 Anomalous Pressures due to Tectonics

Tectonic compression has been invoked as a mechanism for shale overpressuring. This problem might be analysed in terms of an applied volumetric strain rate. If we assume an isothermal situation, then the conditions for overpressure development are

$$\frac{\partial u}{\partial t} > 0 \quad \Rightarrow \quad Q\zeta \frac{\partial \varepsilon}{\partial t} > \nabla\cdot(\underset{\approx}{\mathbf{K}}\, \nabla\cdot\phi^*) \tag{38}$$

where positive strain rate represents volume decrease or compaction. The product $Q\zeta$ is determined to be 12966 MPa and 17285 MPa for the shallow and deep cases, respectively.

With volumetric strain rate negative, signifying volume increase or dilatancy, we would expect underpressure development. However the problem of underpressure development by dilatancy due to tectonic activity is likely to be much more complex than this theory suggests. We can distinguish two types of dilatancy in overconsolidated mudrocks, the first being the (nonlinear) elastic volume increase associated with rebound and the second, the volume increase due to the brittle fracture. Brittle fracture is largely a consequence of shear stresses acting within the material. Whether a mudrock compacts or dilates under high shear stress will depend on the magnitude of the mean normal effective stress. Under conditions of relatively low mean normal effective stress combined with high shear stress there is a strong tendency for a mudrock to dilate and, with restricted groundwater movement, an underpressure situation could develop.

Many regions of Switzerland, including the Jura, are currently tectonically active (SCHAER and JEANRICHARD, 1974; SCHAER et al., 1975). *There is a good possibility that tectonic deformation of the Opalinus Clay influences the present day hydrogeology of the formation.* We note the strong indications of tectonic shearing in the Riniken and Schafisheim boreholes in this context. Quantitative analysis of this problem would require additional development of the theory to incorporate shear stress effects. Estimates would also be required of the rate of deformation and of various strength parameters.

4.8 Conventional Effective Stress

Referring to equation (15) we find that the ratio K/H to be 0.996 and 0.981 for shallow and deep material, respectively. Thus, as a reasonable approximation, we may re-write (15) as

$$\varepsilon = \frac{1}{K} \sigma_m' - \alpha_b (T - T_0) \qquad (38)$$

where σ_m' is the conventional mean normal effective stress. *Thus for the Opalinus Clay we can justifiably describe deformation in terms of conventional effective stress.* This, once again, is a soil-like characteristic. In most hard rocks K/H is significantly less than unity for short-term deformation (NUR and BYERLEE, 1971), typically 0.80 for a limestone, 0.72 for Kayenta sandstone, and 0.24 for Hanford basalt (PALCIAUSKAS and DOMENICO, 1989).

4.9 One-Dimensional Analysis of Transient Flow

The solution of boundary value problems for the general three-dimensional case is mathematically complex and, for the purpose of our scoping calculations, we choose to examine a simplified one-dimensional situation.

Inspecting values assigned to bulk, water and solids compressibility in Section 4.2, we find $\beta > \beta_w > \beta_s$. The transient flow equations can be considerably simplified if we assume the water and solids to be incompressible ($\beta_w = \beta_s = 0$). Additional simplification is obtained if we take the hydraulic conductivity to be independent of direction (scalar) and constant throughout the clay stratum.

Taking u_{ex} as the excess (or deficient) pressure relative to hydrostatic, then for constant temperature (16) becomes

$$\frac{\underset{\approx}{K}}{\gamma_w \, g} \nabla^2 \cdot u_{ex} + \frac{1}{K}\left[\frac{\partial \sigma_m}{\partial t} - \frac{\partial u_{ex}}{\partial t}\right] = 0 \qquad (39)$$

We now examine the one-dimensional situation in which water flow occurs only in the vertical direction. We assume that the clay is laterally-constrained so that the horizontal principal strain increments $\Delta \varepsilon_2$ and $\Delta \varepsilon_3$ are both zero

$$\Delta \varepsilon_{22} = \Delta \varepsilon_{33} = 0 \qquad (40)$$

The volumetric strain increment $\Delta \varepsilon$ is then given by

$$\Delta \varepsilon = \Delta \varepsilon_{11} + \Delta \varepsilon_{22} + \Delta \varepsilon_{33} = \Delta \varepsilon_{11} \qquad (41)$$

where $\Delta \varepsilon_{11}$ is the vertical principal strain increment. The effective stress increments during constrained loading are related by

$$\Delta \sigma_{22}' = \Delta \sigma_{33}' = \Delta \sigma_{11}' \frac{\nu}{(1 - \nu)} \qquad (42)$$

The mean normal effective stress increment therefore becomes

$$\Delta \sigma_m' = \Delta \sigma_{11}' \frac{(1 + \nu)}{3\,(1 - \nu)} \qquad (43)$$

Using (38) and (43) we obtain

$$\frac{\Delta \sigma_{11}'}{\Delta \varepsilon_{11}} \frac{(1 + \nu)}{3\,(1 - \nu)} = \frac{\Delta \sigma_m'}{\Delta \varepsilon} = K \qquad (44)$$

By definition

$$\frac{\Delta \sigma_{11}'}{\Delta \varepsilon_{11}} = E_k \tag{45}$$

Using (43), (44) and (45), the transient flow equation (39) becomes

$$\frac{\partial u_{ex}}{\partial t} = C_v \frac{\partial^2 u_{ex}}{\partial z^2} + \frac{\partial \sigma_v}{\partial t} \tag{46}$$

where C_v is the coefficient of consolidation, given by

$$C_v = \frac{E_k \mathbf{K}_v}{\gamma_w g} \qquad \text{and} \qquad \sigma_v = \sigma_{11} \tag{47}$$

4.9.1 Approximate Duration of Transient Flow

In a low permeability mudrock stratum the duration of the transient flow response produced by a particular geological process may considerably exceed the duration of the process itself. Thus, as a first approximation, we can assume that the geological process occurred instantaneously at some time in the past and, thereafter, the clay remained essentially undisturbed while equilibrium was slowly regained. This simple approach has the great advantage that we can apply a well-known analytical solution for the one-dimensional consolidation problem in our calculations.

Considering a horizontal stratum of clay of thickness $2H$ with overlying and underlying aquifers providing perfect drainage. Assume that at some time $t = 0$ a geological event instantaneously produces an excess (or deficient) pore pressure u_0 in the clay which is uniform throughout the thickness of the stratum. Take z to be the vertical coordinate with the positive direction upwards. If the total stress during the transient flow phase is constant then (46) becomes

$$\frac{\partial u_{ex}}{\partial t} = C_v \frac{\partial^2 u_{ex}}{\partial z^2} \tag{48}$$

which is the basic differential equation of Terzaghi's consolidation theory.

The boundary conditions are

$t = 0,$	$0 \leq z \leq 2H,$	$u_{ex} = u_0$
$0 < t \leq \infty,$	$z = 0,$	$u_{ex} = 0$
$0 < t \leq \infty,$	$z = 2H,$	$u_{ex} = 0$
$t = \infty,$	$0 \leq z \leq 2H,$	$u_{ex} = 0$

Defining the non-dimensional time T_v by

$$T_v = \frac{C_v t}{H^2} \tag{49}$$

the solution $u_{ex} = u_{ex}(z, T_v)$ emerges as a Fourier sine series (TAYLOR, 1948)

$$u_{ex} = \sum_{m=0}^{m=\infty} \frac{2 u_0}{M} \sin \frac{M z}{H} \exp(-M^2 T_v) \tag{50}$$

where $M = (2m + 1)\pi/2$. We note that the solution can be truncated at $m = 4$ without significant loss of accuracy. At any time, the degree of consolidation U_H at the midpoint of the stratum ($z = H$) is defined as

$$U_H = \frac{u_0 - u_{ex}}{u_0} \tag{51}$$

and

$$U_H = 1 - \sum_{m=0}^{m=\infty} \frac{2}{M} \sin M \exp(-M^2 T_v) \tag{52}$$

We now assess the duration of the transient flow phase by calculating the non-dimensional time for the excess pore pressure u_{ex} to fall to 5% of its original value u_0. We find

$$U_H = 0.95 \quad \text{at} \quad T_v = 1.31 \tag{53}$$

Thus from (49) and (53) the elapsed time t_{95} from the geological event is

$$t_{95} = \frac{1.31\, H^2 \gamma_w\, g}{E_k\, \mathbf{K}_v} \tag{54}$$

Re-writing this in a convenient system of units we find

$$t_{95} = \frac{0.42\, H^2 \gamma_w\, g}{E_k\, \mathbf{K}_v} \times 10^{-10} \quad \text{(years)} \tag{55}$$

where H is half the thickness of the stratum (m), γ_w is the density of water ($Mg.m^{-3}$), g is the acceleration due to gravity (9.81 $m.s^{-2}$), E_k is the constrained modulus of elasticity (MPa), and $\mathbf{K}_v$ is the vertical hydraulic conductivity ($m.s^{-1}$).

4.9.2 Finite Difference Solution

Analytical solutions of (46) have been obtained for more realistic stress histories than that considered above, including progressive burial (GIBSON, 1958) and progressive erosion of overburden (KOPPULA, 1983; KOPPULA and MORGENSTERN, 1983). However, approximate solutions are easily obtained using the finite difference method and the effect of any prescribed stress history can be examined by this means.

If we divide the vertical section into equal intervals each of height Δz and we have equal time increments of Δt, then for optimum numerical stability and accuracy

$$\alpha = \frac{C_v\, \Delta t}{(\Delta z)^2} = \frac{1}{6} \tag{56}$$

which allows us to determine the best value for Δt (FORSYTHE and WASOW, 1960).

The recurrence formula is given by

$$u_{i+1,j} = \alpha (u_{i,j-1} + u_{i,j+1}) + (1 - 2 \alpha) u_{i,j} + \sigma_{i+1,j} - \sigma_{i,j} \qquad (57)$$

where i and j are the range variables associated with time and vertical height, respectively. The total vertical stress is calculated from overburden thickness and the latter is incremented or decremented at each time step to simulate deposition or erosion.

4.10 Anomalous Pressures and the Opalinus Clay

The vertical hydraulic conductivity $\mathbf{K_v}$ of intact Opalinus Clay is estimated to lie in the range 10^{-13} to 10^{-15} m.s^{-1}. Although the hydrogeological significance of fractures has yet to be fully quantified, fracture flow is likely to be the dominant mechanism of vertical water movement at the regional scale. Using the "equivalent porous medium" approach we guess $\mathbf{K_{vf}}$ for the fractured clay to lie in the range 10^{-11} to 10^{-13} m.s^{-1}. *Thus our main difficulty in calculating the probable duration of a transient flow episode within the Opalinus Clay is the uncertainty in the selection of an appropriate hydraulic conductivity.* Table 3 shows t_{95} (years) for a wide $\mathbf{K_v}$ range, from 10^{-11} to 10^{-15}, and for E_k values from both the shallow and deep parameter sets. This stratum thickness is taken as 100 m giving $H = 50$ m.

These calculations suggest that a deeply buried stratum of clay-shale will display a hydrogeological transient of shorter duration than a stratum of equal permeability at shallow depth. This effect is entirely due to the predicted increase in constrained modulus E_k, and hence coefficient of consolidation C_v, with depth.

Our first conclusion, assuming the parameter ranges to be appropriate, is that *geological events at times significantly greater than 2 x 10^6 years from the present day are unlikely to be responsible for abnormal pressures in the Opalinus Clay.* We must qualify this finding by noting that the implications of coupled flow to the long-term transient response have yet to be fully explored (see Sections 5.5 and 5.6).

For the fractured clay we calculate generally fairly short t_{95} values in the range 6 x 10^1

to 2 x 10^4 years.

However, in order for the stratum to abstract (or expel) water via the fracture network, there must be an exchange of water between the fractures and the intact clay. The time for such an exchange to take place will depend on on the lengths of the flow paths and will increase with increasing fracture spacing. It seems improbable that equilibrium between the fractures and the intact clay could be established in as little time as 6 x 10^1 years, suggesting that the lower values of t_{95} in Table 3 may be misleading.

Table 3: Elapsed Times for 95% Excess (or Deficient) Pore Pressure Dissipation at the Midpoint of the Opalinus Clay

Vertical Hydraulic Conductivity ($m.s^{-1}$)		Elapsed time t_{95} (years) Shallow	Deep
10^{-11}	Frac.	2 x 10^2	6 x 10^1
10^{-12}	"	2 x 10^3	6 x 10^2
10^{-13}	Frac./Unfrac.	2 x 10^4	6 x 10^3
10^{-14}	Frac.	2 x 10^5	6 x 10^4
10^{-15}	"	2 x 10^6	6 x 10^5

This analysis does not take into account the possible effect of intercalated beds and lenses of sands and siltstones within the clay. Whether such features could act as "drainage layers" is unknown.

The general picture which emerges from these calculations is one in which fracture flow may fairly rapidly re-equilibriate after the geological event but re-adjustments of the water content of the intact clay may occur over a much longer time period. *Thus fracture flow might be regarded as being essentially out of phase with flow in the intact clay.* For the case where erosion is a source of underpressure, the intact material must draw in water from the fracture network. In the early stages of this process we would anticipate

localized swelling in the vicinity of the fractures as depicted in Fig. 6.

Given that we have excluded geological processes which occurred at times greater than 2×10^6 years from the present, *we identify erosion (glacial and/or fluvial) as a possible source of abnormal pressuring*. In the Homburger Tal region up to 350 m of overburden may have been removed by valley erosion. We model this situation using the finite difference method outlined above. Assuming the duration of this process to be 10^6 years, the average rate of erosion is 3.5×10^{-4} m.year^{-1}. Taking the total stress gradient to be 0.025 MPa.m^{-1}, the rate of change of total stress is 8.75×10^{-6} MPa.year^{-1}.

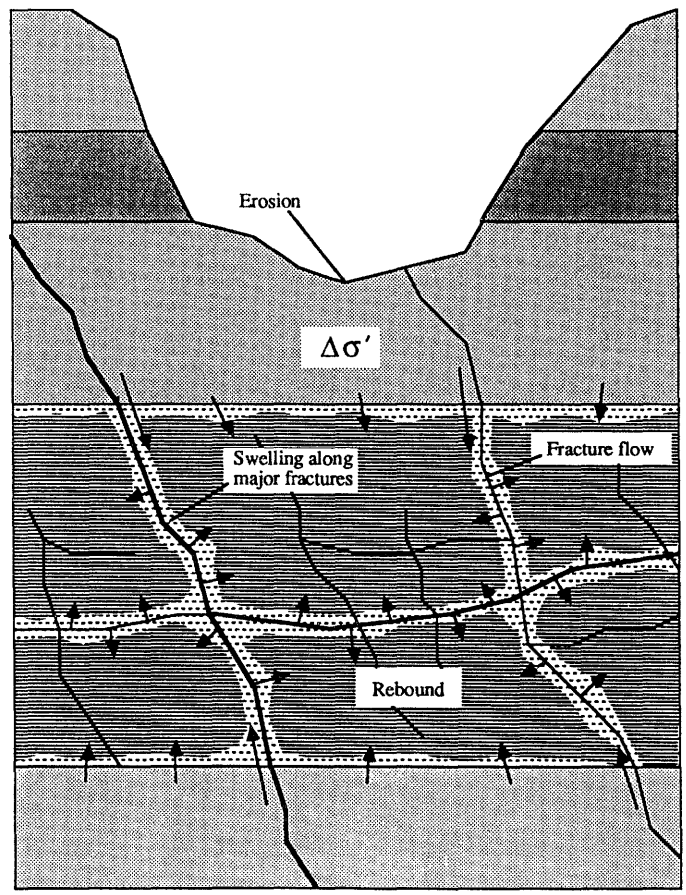


Fig. 6: Schematic of the possible hydrogeological effects within a mudrock stratum of simple rebound associated with overconsolidation. Valley erosion produces a change in vertical stress on the mudrock resulting in an "underpressure type" hydraulic transient. Water is drawn in through the fracture network producing swelling in the vicinity of the fractures.

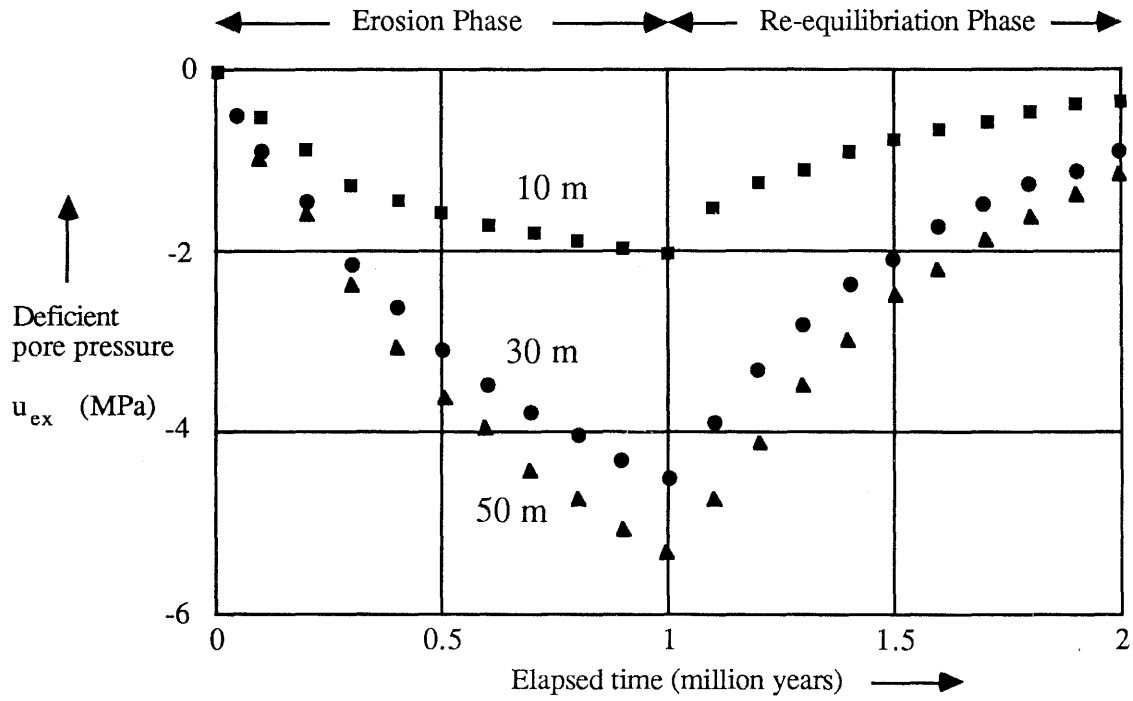


Fig. 7: Deficient pore pressures in a 100 m thick mudrock stratum subject to stress changes associated with the erosion of a 350 m deep valley. Pressures are plotted against time at points 10, 30 and 50 m into the stratum. Assumed parameters are $\mathbf{K}_v = 10^{-15}$ m.s^{-1} and $E_k = 504$ MPa.

Fig. 7 *illustrates* the response to erosion and shows the deficient pore pressures at 10, 30 and 50m into the clay stratum plotted against time for $\mathbf{K}_v = 10^{-15}$ m.s^{-1} and $E_k = 504$ MPa. Given the large possible range for $\mathbf{K}_v$, the quantitative approach cannot sensibly be persued beyond this point.

Finally, we cannot discount the possibility that neotectonic activity and the associated uplift, subsidence or tilt of the ground surface can produce hydrogeological anomalies similar to the ones described above. Precise levelling in the Alps suggests that present

day rates of deformation are similar to those which have been occurring for the last 10 - 20 million years (SCHAER and JEANRICHARD, 1974; SCHAER, 1979). During this century, the Alps have undergone uplift while the Jura (Basel area) has shown a downward movement.

5 COUPLED FLOW

We now move on to examine some of the implications of coupled flow to groundwater movement in mudrock environments.

In general terms a coupled flow process is one in which flow of any kind (e.g., fluid, solute, heat, electrical current) is driven by the gradient of a potential not usually associated with that flow or, in mathematical terms, the flow depends on a non-conjugate force.

The flows J_i are usually assumed to be linearly related to the gradients X_i as follows:

$$J_i = -\sum_{k=1}^{n} L_{ik} X_k \qquad i = 1, 2, \ldots\ldots n \tag{58}$$

where the subscripts i and k indicate the various types of flows and driving gradients, respectively, and L_{ik} are their phenomenological coefficients (KATCHALSKY and KURRAN, 1967; BEAR, 1972). Onsager's reciprocal relation suggests that the coefficients are related as follows:

$$L_{ik} = L_{ki} \tag{59}$$

Non-hydraulically driven flow is given the general name osmosis. The term chemico-osmosis refers to flow driven by a chemical concentration gradient, thermo-osmosis to flow driven by a temperature gradient and electro-osmosis to flow driven by an electrical potential gradient (strictly flow is driven by the *negative* gradient).

Fig. 8 illustrates direct (on-diagonal) and coupled (off-diagonal) flow phenomena.

Electro-osmosis has been used as a method of de-watering in civil engineering excavations in clay and is currently under investigation as a technique for controlling the movement of chemical pollutants. Thermo-osmosis is likely to be a mechanism of water movement around a repository containing heat-emitting wastes. Numerical simulations of coupled transport processes, including thermo- and chemico-osmosis, in saturated clays have been reported by CARNAHAN (1985; 1989).

We identify chemico-osmosis as a first-order effect, and electro- and thermo-osmosis as second-order effects in natural mudrock environments. Only chemico-osmosis will be considered further in this report, and for the sake of brevity we will refer to this process simply as "osmosis".

	Gradient X			
Flow J	Hydraulic Head	Temperature	Electrical	Chemical Concentration
Fluid	**Advection** *Darcy's Law*	Thermo-osmosis	Electro-osmosis	Chemico-osmosis
Heat	Isothermal heat transfer	**Thermal conduction** *Fourier's Law*	Peltier effect	Dufour effect
Current	Streaming current	Thermo-electricity	**Electrical conduction** *Ohm's Law*	Diffusion & membrane potentials
Ion	Streaming current	Soret effect (thermal diffusion of electrolyte)	Electro-phoresis	**Diffusion** *Fick's Law*

Fig. 8: Coupled and direct flow phenomena relating to the movement of fluid, heat, electrical current and ions. Direct flow mechanisms are shown in bold typeface and appear on the main diagonal. The associated physical law is given in italics. Coupled mechanisms lie off the main diagonal (adapted from: MITCHELL, 1976).

5.1 Clays as Osmotic Membranes

An ideal osmotic membrane is a semi-permeable barrier that allows transport of the solvent but not of the solutes. Solutes are rejected on the basis of size and/or electrical restrictions. Clays can act as membranes of varying degrees of ideality, an attribute which is due to electrical restrictions on anion movement within the adsorbed water. The negative charged surface of a clay platelet and its cation-dominated sorption layer together make up the Gouy diffuse double layer (GOUY, 1910; 1917). In compacted clays the clay platelets are forced into close proximity so that the double layers interact. The inter-platelet space has a negative electrical potential which repels anions attempting to migrate through this space. In order to maintain electrical neutrality in the outer or "free solution", cations must remain with their counter-ions and their movement through the clay is also restricted (FRITZ, 1986).

5.2 Experimental Studies of Coupled Flow

Much experimental work has been undertaken on coupled flow processes (OLSEN, 1969; 1972), aspects of membrane composition, fluid composition, membrane thickness, overall membrane efficiencies (FRITZ and MARINE, 1983), and the effects of experimental compaction pressures and temperatures on solute mobility. The possibility that osmotic phenomena might account for deviations from Darcy's Law has also been examined (MITCHELL and YOUNGER, 1967; OLSEN, 1965; 1985). Hyperfiltration (reverse osmosis) has also been investigated as a mechanism for the precipitation of calcite cements (FRITZ and EADY, 1985).

Most experimental studies have been performed on pure, artificially compacted clays and very little laboratory research has been undertaken on natural samples.

5.3 Osmotic Pressure across an Ideal Membrane

The osmotic pressure $\Delta\Pi$ that can be generated across an ideal membrane can be calculated from the solution properties on either side of the membrane

$$\Delta \Pi = \frac{\mathbf{R}\,T}{V_w} \ln\left[\frac{a_A}{a_B}\right] \qquad (60)$$

where **R** is the gas constant (0.0082 MPa.l.mol^{-1}.°K^{-1}), T is the absolute temperature, V_w is the mean of the partial molar volumes of water on either side of the membrane (l.mol^{-1}), and a_A and a_B are the activities of the water (FRITZ and MARINE, 1983). The molar volume of pure water is 0.018 l.mol^{-1} at 25°C.

The activity of the water is calculated from the approximation

$$a = 1 - 0.017 \sum m_i \qquad (61)$$

where m_i are the molalities of dissolved anions, cations and neutral species (GARRELS and CHRIST, 1965).

An approximate calculation for $\Delta\Pi$ is given by the the van't Hoff equation

$$\Delta\Pi = \eta \mathbf{R} T \left[C_B - C_A \right] \qquad (62)$$

where η is the number of constituent ions of the dissociating solute ($\eta = 2$ for NaCl and $CaSO_4$ and $\eta = 3$ for Na_2SO_4 and $CaCl_2$) and C_A and C_B are the solute concentrations expressed in molarity. For 1:1 electrolytes like NaCl the pressure predicted by (62) is within 5% of that given by (60) if the concentration difference across the membrane is less than 1 mol.l^{-1} (FRITZ, 1986). We use (62) exclusively in this report.

We now perform a number of simple scoping calculations for the Opalinus Clay and underlying sediments.

For the NaCl pore waters of the Liassic/Keuper underlying the Opalinus Clay we take C_B = 1.28 mol.l^{-1}. Assuming a temperature of 30°C, we estimate $\Delta\Pi$ with respect to fresh water ($C_A = 0$) to be 6.4 MPa.

If we assume the results of pore water squeezing experiments to be indicative of the pore water chemistry of the Opalinus Clay, then C_B = 0.28 mol.l^{-1} NaCl. Taking a temperature of 30°C as before, we calculate $\Delta\Pi$ with respect to fresh water to be 1.4 MPa.

Thus, assuming an ideal membrane, osmotic pressure differential between the Liassic/Keuper and the Opalinus Clay is calculated to be approximately 5 MPa, *suggesting* a downward osmotically-driven flux from the clay to the underlying formations.

The water chemistry of the clay is of course highly uncertain. Precisely what *fraction of the total water is expelled* during squeezing experiments is unknown, as is the *distribution* of this water between the micro- and macroporosity. It is known, however, that squeezing experiments (up to 50 MPa) generally yield higher concentrations than leaching experiments and that, if the residues from squeezing are subsequently leached, the resulting concentration is lower than that obtained by the exclusive use of squeezing or leaching. Fresh samples also tend to give higher concentrations than experiments on older, and possibly more oxidised, material (ROSS, pers. comm., 1989).

It should also be noted that the pore water salinity of the formations underlying the Opalinus Clay is based on the analysis of water samples from oil wells some distance away from the clay-sampling locations.

5.4 Non-Ideal Membranes

In non-ideal membranes the solute diffuses through the membrane in a direction which is opposite to that of the osmotically-driven flux of water. The differential pressure is then less than that predicted by (60) or (62). The reflection coefficient σ is defined as the ratio of the observed osmotic pressure ΔP to its theoretical maximum $\Delta\Pi$ when the net flux of solution J_w across the membrane is zero (STAVERMAN, 1952),

$$\sigma = \left[\frac{\Delta P}{\Delta \Pi}\right]_{J_w = 0} \qquad (63)$$

MARINE and FRITZ (1981) developed a model which describes σ in terms of the porosity and surface charge density of a clay membrane and the mean solute concentration on either side. The model suggests that σ is highly dependent on the degree of compaction and the cation exchange capacity of the membrane. For equal porosities, a high CEC clay such as smectite is likely to exhibit higher σ than a lower

CEC clay such as illite. As porosity decreases to zero, σ approaches unity for the clay minerals examined. Experiments on sodium bentonite by FRITZ and MARINE (1983) confirm this trend.

For Opalinus clay with a porosity in the range 8 - 12% and a total CEC of roughly 25 meq/100g, we anticipate σ to be moderately high and in the range 0.5 - 0.8 for typical water chemistries.

5.5 Transient Flow under Combined Hydraulic and Chemical Gradients

A theory of one-dimensional transient flow in a non-ideal clay membrane under combined hydraulic and chemical gradients has been presented by GREENBERG (1971) and MITCHELL et al. (1973).

An isotropic, homogeneous, horizontal layer of saturated clay is considered under isothermal conditions. It is assumed that no ion exchange or adsorption reactions occur during diffusion and that the total stress remains constant throughout the transient flow period. From the postulates of irreversible thermodynamics, equations for the flow of water and for the flow of solute in the water can be derived (KATCHALSKY and CURRAN, 1967; LETEY and KEMPER, 1969).

By assuming the compression behaviour of the clay to be linear, GREENBERG (1971) obtained the following (see Appendix B)

$$\frac{1}{C_v}\frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial z^2} + \frac{\mathbf{K}_{hc}\,\gamma_w\, g}{\mathbf{K}} \cdot \frac{\partial^2 C_s}{\partial z^2} \qquad (64)$$

and

$$n\frac{\partial C_s}{\partial t} = \frac{\boldsymbol{K}_{ch}}{\gamma_w\, g} \cdot \frac{\partial}{\partial z}\left((C_s/C_{sm})\frac{\partial u}{\partial z}\right) + \mathbf{D}'\frac{\partial^2 C_s}{\partial z^2} - \frac{C_s}{E_k}\frac{\partial u}{\partial t} \qquad (65)$$

where u is the pore pressure, n is porosity, C_s is the solute concentration (mol.m^{-3}), C_{sm} is the maximum value of C_s, E_k is the constrained modulus of elasticity, C_v is the coefficient of consolidation and

$$K_{ch} = \mathbf{K}_{ch} + C_{sm}\,\mathbf{K} \tag{66}$$

$$\mathbf{D}' = \mathbf{D} + C_s\,\mathbf{K}_{hc} \tag{67}$$

where **K** is the hydraulic conductivity ($m.s^{-1}$), **D** is the diffusion coefficient for the solute ($m^2.s^{-1}$), and $\mathbf{K}_{ch}$ ($mol.s^{-1}.m^{-2}$) and $\mathbf{K}_{hc}$ ($m^5.mol^{-1}.s^{-1}$) are coupling coefficients.

In the absence of coupling $\mathbf{K}_{ch} = \mathbf{K}_{hc} = 0$ and (64) reduces to (48), the Terzaghi equation for one-dimensional consolidation. For no coupling and no hydraulic gradient (65) reduces to the solute diffusion equation derived from Fick's law.

Three types of coupling are indicated by (64) and (65). Chemico-osmotic coupling, or the movement of water due to a solute concentration gradient is given by the second term on the right hand side of (64). The first term on the right of (65) describes solute flow under a hydraulic gradient. The third term on the right of (65) relates to solute movement due to porosity changes ("mathematical" rather than "Onsagerian" coupling).

This theory is based on the concept of the simple porous medium and, since no distinction is made between free and adsorbed water, the solute concentration of the pore water C_S cannot be precisely defined. Double layer theory suggests that solute concentration in a clay is highly heterogeneous at the microscopic scale.

5.6 Long-term Transients and Coupled Flow

In Chapter 4 we assumed that long-term transient flow is exclusively driven by hydraulic gradients. If we acknowledge osmotically-driven flow to be a significant component of the total flow in mudrocks, then *long-term transients must be viewed as the response to both hydraulic and chemical disequilibrium* in the geological system.

If we assumed total stress to be constant during transient flow, then we could explore the coupled flow phenomenon using a finite difference solution of (64) and (65). Since clay-water interaction is not explicitly examined in the theory of transient flow, the validity of this approach remains open to question.

We see the further development and application of the equations of transient flow under

combined hydraulic and chemical gradients in mudrocks subject to varying total stress as a major priority.

5.7 Coupled Flow in Sedimentary Basins

ALEXANDER (1990) outlines osmosis and its potential for modifying *in situ* the direction and rate of groundwater flow (hence groundwater head variations) and associated solute migration. The consequences of the coupling of chemical and hydrogeological processes in a natural system, where ion-exchange, osmosis and reverse osmosis are viewed as distinct processes, are considered.

Coupled flow has been invoked as a mechanism to account for two main types of phenomena observable in sedimentary basins (DE SITTER, 1947; BERRY, 1969; BREDERHOEFT et al., 1963; GRAF et al., 1966; HITCHON et al.,1971; amongst others). These are the occurrence of anomalously high and low hydraulic heads (JONES, 1968), and the existence of fluids of anomalous chemical composition (BREDERHOEFT et al., 1963; GRAF, 1982).

In most aquifer systems the potentiometric surface slopes gently away from the recharge areas towards discharge areas. Consequently potentiometric surface elevations higher than the elevation of recharge areas (JONES, 1968), or lower than those of discharge areas (BERRY and HANSHAW, 1960; BELITZ, 1985), are anomalies difficult to explain by normal gravitational flow of water. Also, if a hydraulic pressure gradient is balanced by an osmotic pressure then there is no net force to cause unidirectional advection. If this situation exists, water movement will not occur even in cases where e permeability and hydraulic gradient indicate that it should. We examine this case further in Section 6.3. Where geological membranes separate solutions of substantially differing concentrations, the resultant osmotic force is capable of generating fluid pressures which approach that of lithostatic pressure (HAYDON and GRAF, 1986).

The possible role of osmosis in groundwater flow in the mixed sedimentary formations beneath Harwell Laboratory in Oxfordshire has been examined by BRIGHTMAN et al. (1987) and ALEXANDER (1990).

6 MEASUREMENT OF HYDROGEOLOGICAL PARAMETERS

In this chapter we examine the implications of coupled flow, borehole effects and swelling on hydrogeological testing in clays. Although much of our discussion relates to the general problem of testing in clays and shales, we do address one specific set of observations which have been made in the Opalinus Clay in the area of the proposed Wisenberg Tunnel in the Homburger Tal. Interpretation of the head measurements in one of the boreholes, RB26B, has proved to be problematic. We explore the possibility that anomalously low hydraulic heads at various depths in this borehole are a consequence of measurement difficulties associated with this particular completion or, alternatively, are symptomatic of processes which may have far-reaching implications to both the testing and hydrogeological modelling of the Opalinus Clay.

6.1 Typical Measurement Systems and Procedures

The difficulties associated with *in situ* hydraulic testing in mudrocks and the lack, in the past, of incentives to examine these rock-types in detail have combined to deter much development in this area. Head measurements in mudrocks are not common practice. Where *in situ* testing has been performed, the testing systems employed have been either (a) standard oil- or water-well single or straddle packer systems operated in open-hole or perforated casing, (b) permanent hydrogeological completions using gravel packs, or (c) soil mechanics completions using piezometers.

In very stable boreholes, single or straddle packer systems can be used to isolate specific sections of interest. These sections are usually cleaned before testing or are drilled with fresh water to minimise wall effects such as smearing. Short duration constant head or pulse tests are used to determine the permeability of the rock. Positive and negative pressures, related to some assumed head, are applied to the test zone. The "true" head is obtained by analysis of the recovery trend. Similar techniques have been applied in cased boreholes by explosively perforating the casing in the zones of interest and running a straddle packer inside the casing (BRIGHTMAN et. al., 1987).

In low permeability clay and shale formations, the extended duration of the flow transients during testing and potential borehole stability problems may prompt the use of

permanent hydrogeological completions. ROBINS et al. (1981) describe a number of gravel pack completions in deep boreholes drilled in a mixed sedimentary sequence at Harwell Laboratory in Oxfordshire. A cavity to accommodate the gravel pack was developed beneath the final casing in each hole by under-reaming. The final completion string, incorporating the gravel pack and well-screen, was lowered into each hole on drill pipe. The packer was inflated inside the casing and 20/40 mesh round gravel was pumped into the annulus around the screen. A second packer, with a plastic pipe extending to surface, was installed in the casing to establish a test zone. Additional details and test results are given in ALEXANDER and HOLMES (1983).

For shallow depths, soil mechanics methods and hardware are applicable (see HANNA, 1973). Piezometer types are distinguished primarily by their "response times" which are a function of quantity of water that must move in from the formation to enable them to operate. The standpipe piezometer consists of a tube with a porous filter element on the end that can be sealed into the ground at the appropriate level. Two types of filter element are generally used, high air entry and low air entry. The Casagrande-type device is the most frequently installed standpipe piezometer. It has a cylindrical (low air entry) porous element protected by a perforated rigid sheath. The response time of this type of piezometer is comparatively slow. Water level in the open standpipe is measured with a dipmeter. In the closed-hydraulic piezometer the groundwater pressure is detected in a small piezometer tip and conducted through narrow-bore plastic tubes to a remote point, where the pressure is measured using a mercury manometer, Bourdon gauge or pressure transducer. These tubes must be regularly de-aired to avoid erroneous results. The closed-hydraulic piezometer has a shorter response time than open-tube systems. Electrical piezometers have a pressure transducer located close to the porous element and, because of their low system compliance, very rapid response is possible with these devices.

Piezometer tips are located in sand packs or filters with a particle size distribution designed to prevent the inward movement of fines from the formation. Bentonite is most often used to provide a seal above the pack, and if the piezometer is not near the base of the borehole, a bentonite seal is also placed beneath the sand pack. The length of the seal is typically 0.5 - 1.5 m. The seals may be emplaced in the form of hand-moulded balls made from bentonite powder and water. Alternatively bentonite pellets may be used provided sufficient time is allowed for the pellets to swell before backfilling the hole. Multiple piezometers may be installed in a single hole provided

that care is taken to achieve proper sealing. The remaining sections of the borehole are filled with a bentonite / cement grout with a permeability the same as, or less than, that of the formation.

BRONDERS (1989) describes a technique being developed by SCK/CEN in Belgium for determining the macropermeability of Boom Clay. A 25m deep borehole, 0.15m in diameter, will be drilled from an underground location within the experimental gallery of the Project Hades URL. A 15m long sintered stainless steel (inox) filter will be installed in the test zone at the bottom of the hole and the upper part will be cased-off. The test zone will be sealed using an inflatable packer inside the casing. Two 26m deep monitoring boreholes will be drilled at distances of 0.5m and 1m from the first hole, and each will be equipped with piezometers. The test zone will be subject to a complex pressure history and interpretation will follow the general scheme of BREDERHOEFT and PAPADOPULOS (1980).

6.2 Measurements in the Area of the Proposed Wisenberg Tunnel

As part of the geological investigations for the proposed Wisenberg Tunnel, a suite of boreholes designated as RB1 to RB30 were drilled by the Swiss Federal Railway Company along the route of the tunnel. Several of the boreholes were completed for long-term hydrogeological measurements and one of these, RB26B, penetrated the Opalinus Clay.

The boreholes were drilling using rotary methods using water taken from the Homburgerbach stream with 1% Antisol (carboxymethylcellulose) additive. The drilling fluid was kept in closed circulation. Prior to instrumentation, the holes were opened and filled to surface with drilling fluid.

Pressure transducers were installed in gravel packs in the uncased section of each borehole. Each pack was saturated in fresh water, bentonite pellets were added to form a sealing layer, and a cement plug, typically 11 m in length, was poured to effectively seal-off the test section. Four of these instrumented packs were installed in each borehole (Fig. 9). Electrical wiring was taken through the cement plugs to the surface readout instruments. Table 4 gives completion details for borehole RB26B (NAGRA, pers. comm., 1989).

In borehole RB26B, after the drilling phase and before transducer installation and final completion, caving of the uppermost weathered part of the clay made it necessary to re-drill to 45 m below the surface and install casing to 22 m.

Table 4: Completion Details for Borehole RB26B

Transducer	Depth (m.b.g.s.)	Gravel pack depth interval (m.b.g.s.)	First intercepted (date)	Transducer installed (date)
PZ6	24.0	26.0 - 22.0	28.03.1988	22.04.1988
PZ7	40.0	42.2 - 37.0	30.03.1988	22.04.1988
PZ8	59.8	63.0 - 58.0	31.03.1988	21.04.1988
PZ5	80.0	92.0 - 74.0	06.04.1988	21.04.1988

The total head versus time plots constructed for the four measuring points in RB26B exhibit a rapid *decline* over the first four weeks from the start of measurement (Fig. 10). With the surface as datum (z = 0) for the elevation head, the total head **h** at the end of this initial period is -24 m for PZ6 (24.0 m.b.g.s.), -40 m for PZ7 (40.0 m.b.g.s.), -56 m for PZ8 (59.8 m.b.g.s.) and -76 m for PZ5 (80.0 m.b.g.s.).

After this initial decline, the responses of the four transducers differ somewhat one from another, but the general pattern is an increase in **h** over the next year or so of monitoring (Fig.11). This increase is most apparent for PZ6, the shallowest transducer, which asymtotically approaches **h** = -12 m. The response of PZ7 is similar to PZ6, but less pronounced, with **h** approaching -37 m. PZ8 is fairly constant throughout the monitoring period with **h** = -58 m after a year. Finally PZ5 remains fairly constant for 30 weeks or so, then rises gradually to -71 m, showing no signs of stabilising by then end of the monitoring period.

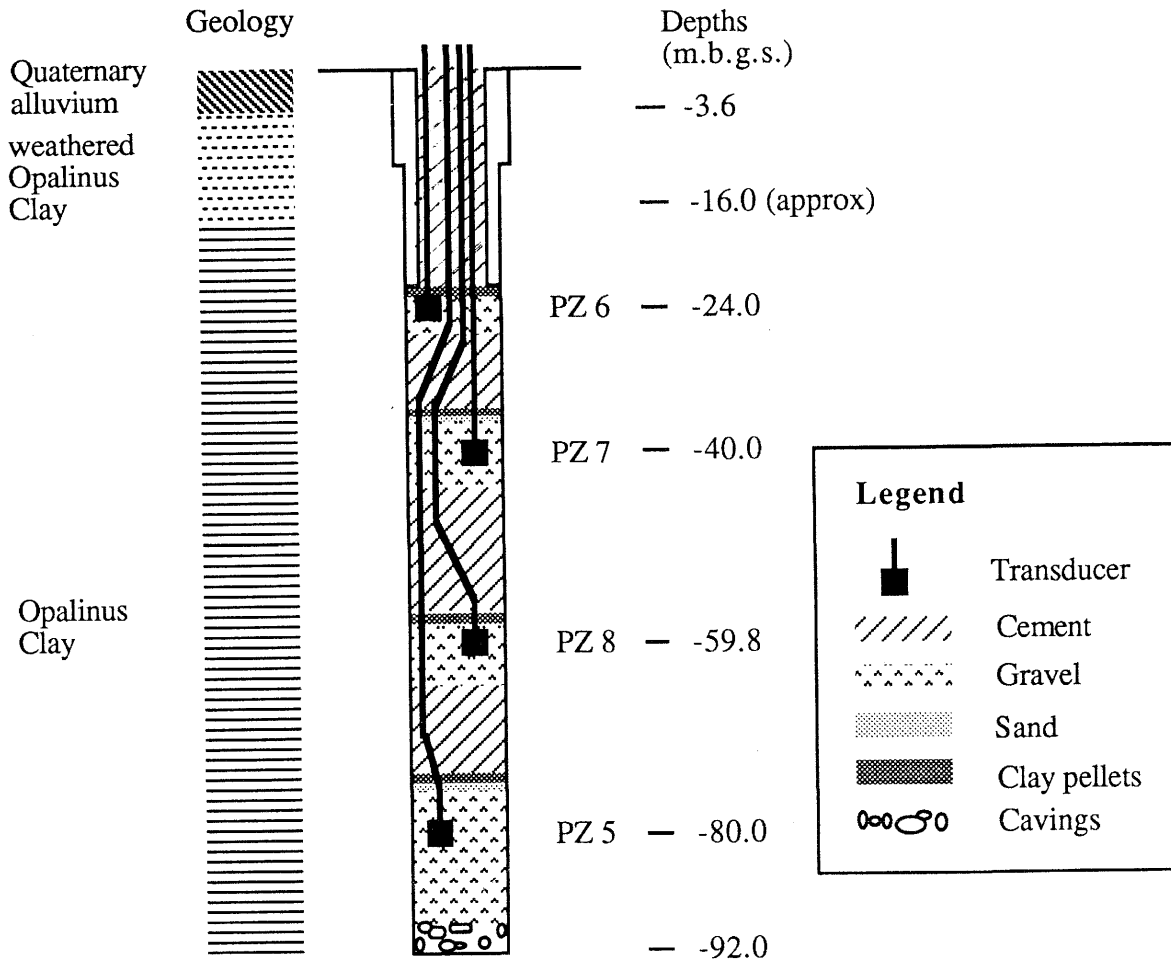


Fig. 9: Hydrogeological completion of Borehole RB26B at the site of the proposed Wisenberg Tunnel, Homburger Tal.

Measurements over the same period indicate that the water table was within 1.4 m of the surface. The hydraulic conductivity of the clay-shale in the vicinity of PZ7 and PZ8 is reported to be $< 10^{-12}$ m.s^{-1} (below measuring limits).

The pressure head h_p at the end of the initial period of sharp decline in RB26B is close to zero at each of the four transducer elevations, and that the water pressures are close to

atmospheric (subnormal). Since the test water will tend to cavitate at high suctions and the pressure transducers probably cannot respond to negative pressures, it is possible that the initial response is more severe than the measurements reveal.

The head reponse in RB26B is in marked contrast to that shown in RB25B, an adjacent hole which penetrates a mixed sequence of limestones, marls and clay-shales (Lower Dogger - Blagdeni, Humphriesi, Sowerbyi and Murchinae Beds) with typical hydraulic conductivity in the range 5×10^{-11} to 1×10^{-12} $m.s^{-1}$. The initial response is a sharp *increase* in **h** which persists for a number of weeks and is followed by more gradual build-up over the next year or so. At the end of the measurement period (67 weeks from start), the total heads **h** are +7 m for PZ4 (24.0 m.b.g.s.), +15 m for PZ3 (36.0 m.b.g.s.), +30 m for PZ2 (50.0 m.b.g.s.) and +4 m for PZ1 (75.0 m.b.g.s.). The data suggest a large hydraulic gradient between PZ2 and PZ1. The important point to emphasise is that, in contrast with RB26B, all total heads measured in RB25B are *positive* (w.r.t. a surface datum). Surface elevations at RB25B and RB26B are 441.00 m and 440.90 m above mean sea level, respectively.

The question raised is "Why should the measured total heads **h** in a borehole in the Opalinus Clay be significantly negative (with respect to a surface datum)?".

Although it is not possible to totally exclude *simple explanations* for the response of RB26B, such as the presence of air in the test sections or the imbibing of water by the bentonite seal, since both RB25B and RB26B were completed in the same way, it is difficult to invoke a completion-related mechanism which would not effect both holes in a similar way. Could this therefore be a particular feature of hydrogeological testing within the clay-shales of the Opalinus Clay? We will explore this possibility later in this chapter.

6.3 Implications of Coupled Flow in Hydrogeological Testing

Osmotically-driven flow has profound implications to the interpretation of hydrogeological tests in compacted clays and mudrocks.

In Section 3.2 we noted that the total head **h** in a clay exhibiting chemico-osmotic effects *may be considered* to have three components

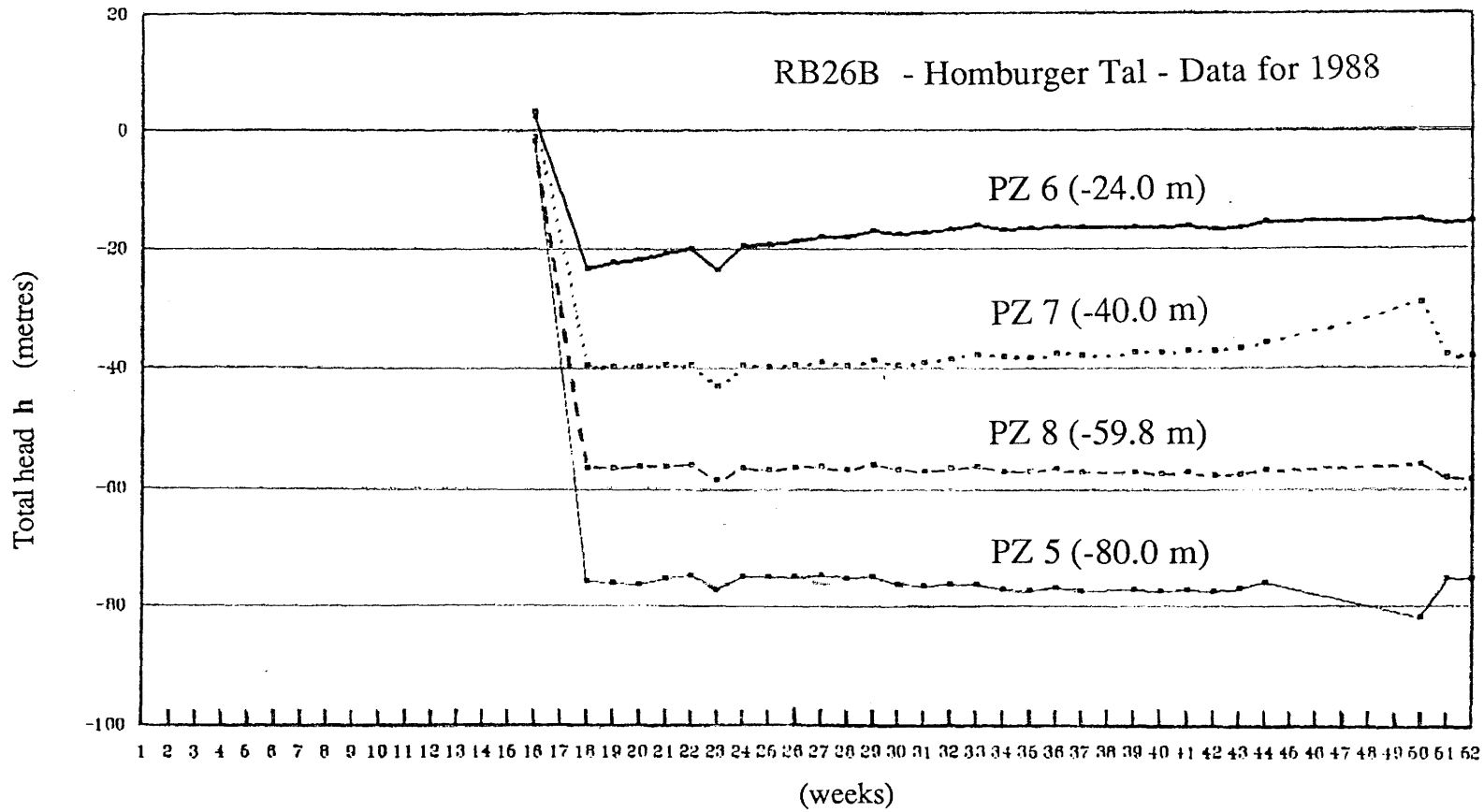


Fig. 10: Head measurements in Borehole RB 26B, site of the proposed Wisenberg Tunnel: Data for 1988. Total head **h** is depicted, with datum at ground surface (440.90 metres above mean sea level).

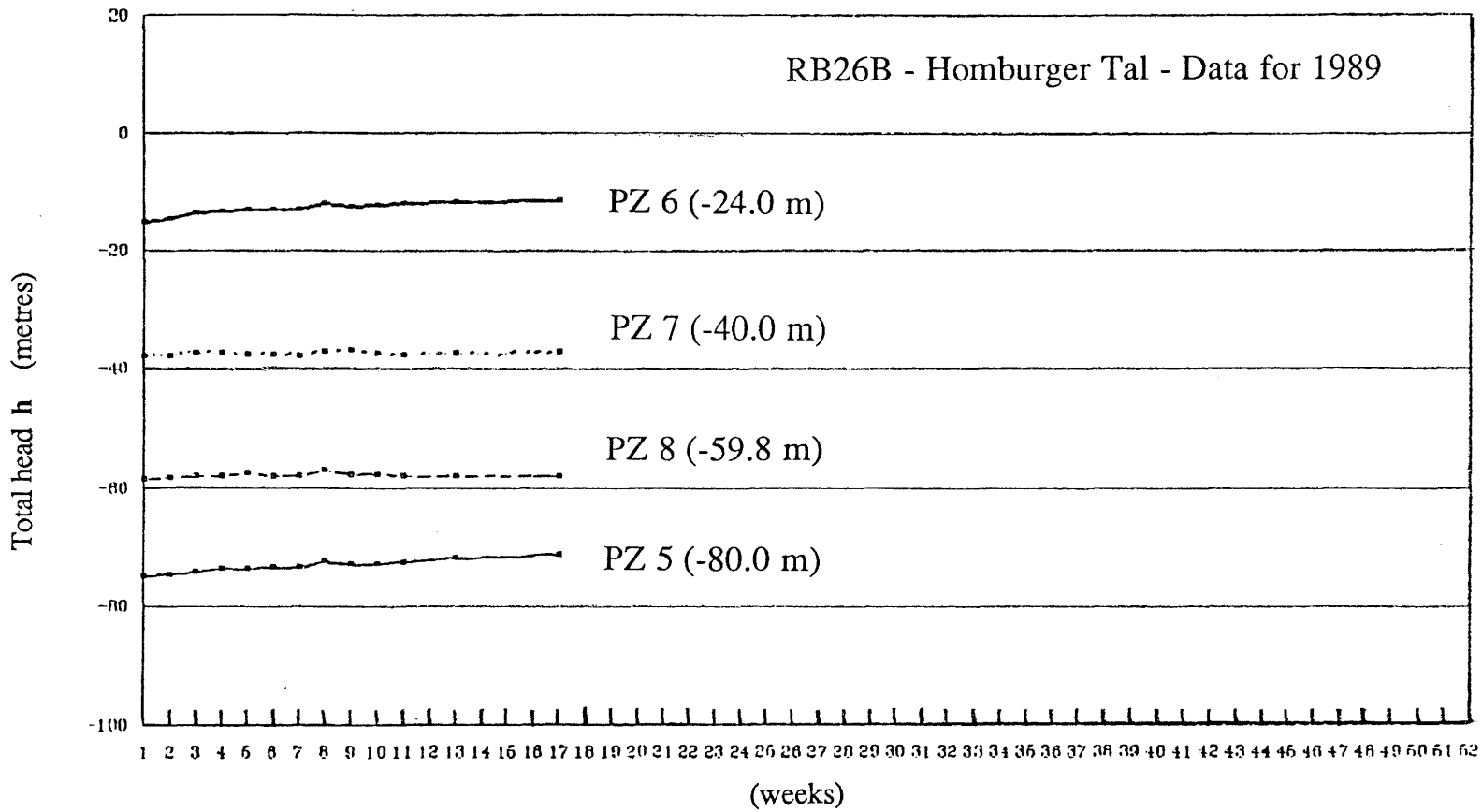


Fig. 11: Head measurements in Borehole RB 26B, site of the proposed Wisenberg Tunnel: Data for 1989. Total head **h** is depicted, with datum at ground surface (440.90 metres above mean sea level).

$$\mathbf{h} = z + h_p + h_s \tag{68}$$

where z is the elevation head, h_p is the pressure head and h_s is the solute head.

Referring to Fig. 12, consider a piezometer containing pure water installed in a stratum of clay. Assume that the advective flux is at all points zero (i.e. the system is in equilibrium). Points 0, 1, 2 and 3 are all at the same elevation as the datum (z = 0). Point 0 is inside the piezometer tip in pure water and the total head $\mathbf{h}_0$ at this point is

$$\mathbf{h}_0 = 0 + h_{p0} + 0 \tag{69}$$

and the water pressure u_0 is

$$u_0 = \gamma_w g h_{p0} \tag{70}$$

Points 1, 2 and 3 are all within the clay stratum and since the electrolyte concentration at all these points is finite

$$\begin{aligned} \mathbf{h}_1 &= 0 + h_{p1} + h_{s1} \\ \mathbf{h}_2 &= 0 + h_{p2} + h_{s2} \\ \mathbf{h}_3 &= 0 + h_{p3} + h_{s3} \end{aligned} \tag{71}$$

Double layer theory (Appendix C) suggests that the electrolyte concentration at point 1, which is close to a clay platelet, will exceed that at point 2, which is midway between two clay platelets, which in turn will grossly exceed that of point 3 which is in a macropore. Thus, bearing in mind the sign of the solute heads,

$$h_{s1} < h_{s2} << h_{s3} < h_{s0} = 0 \tag{72}$$

Now, the condition of zero advective flow requires that $\mathbf{h}_0 = \mathbf{h}_1 = \mathbf{h}_2 = \mathbf{h}_3$, which gives

$$h_{p1} > h_{p2} >> h_{p3} > h_{p0} \tag{73}$$

and

$$u_1 > u_2 >> u_3 > u_0 \tag{74}$$

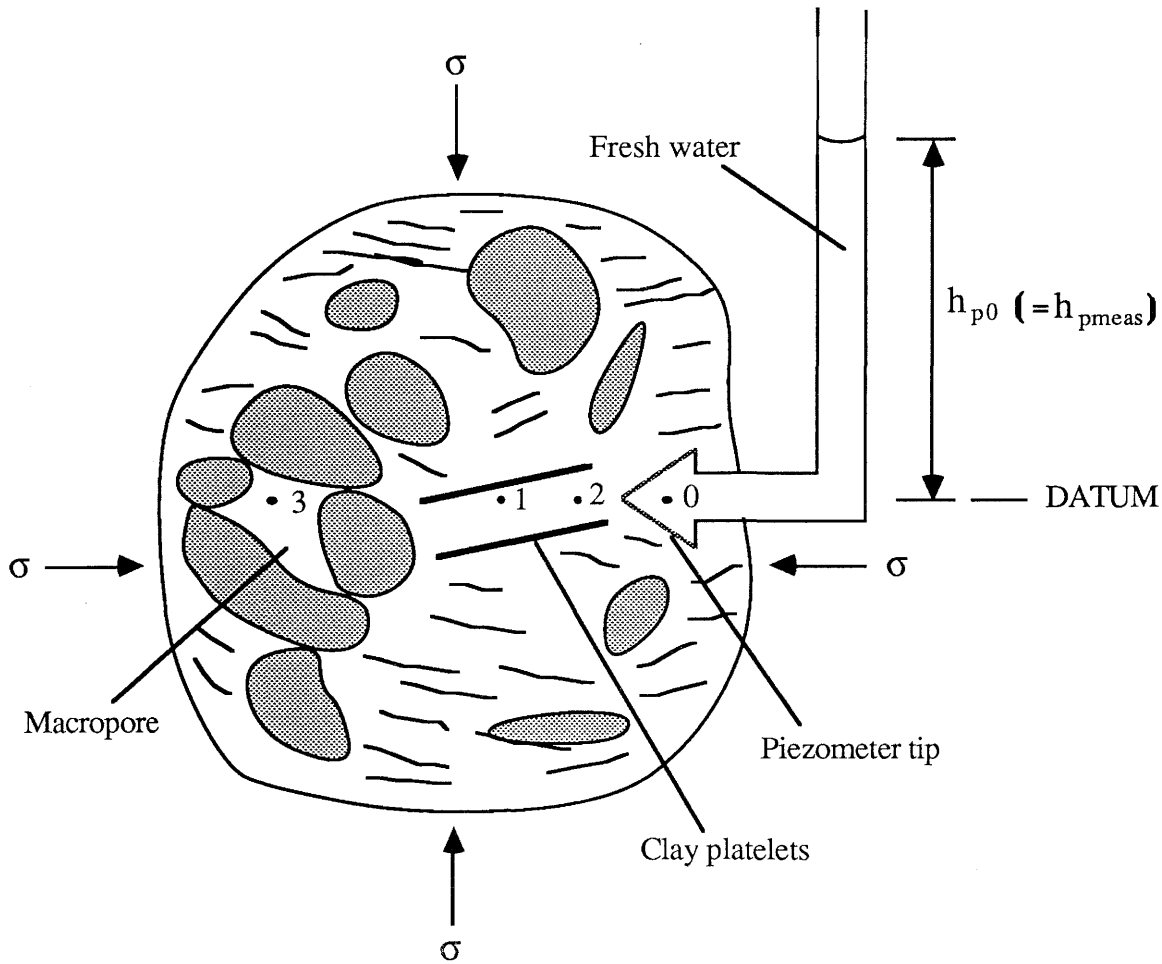


Fig. 12: Diagram illustrating local variation of the pressure head and solute head components of total head in a mudrock and the relationship between measured pressure head and the in situ parameters (adapted from MITCHELL, 1976).

Thus, we reach the important conclusion that, in a clay at equilibrium, the pressure heads and solute heads must vary locally at the microscopic scale. Furthermore, *a piezometer filled with pure water registers a pressure head* h_{p0} *which is equivalent to the sum of the pressure heads and solute heads at all points in the clay which are at the same elevation* (MITCHELL, 1976), or

$$h_{p0} = h_{p1} + h_{s1} = h_{p2} + h_{s2} = \ldots\ldots \qquad (75)$$

Thus, since osmotic pressures are considered to be a source of long-range repulsions due to double layer interactions, *measured pore water pressures in clays may include contributions from long-range interparticle forces* (BOLT and MILLER, 1958). This finding has considerable repercussions when we attempt to establish the exact meaning of the term "effective stress" in compacted clays (see Section 6.4.1).

We now move on to examine the broader significance of these findings. Referring to Fig. 13, consider a thick, horizontally-bedded stratum of clay with an overlying fresh water aquifer and an underlying saline aquifer. Assume that the system is in equilibrium, that is, the total head gradient is at all places zero and that the clay acts as an ideal osmotic membrane. Furthermore, assume that the datum for the elevation head is at the surface, which also corresponds with the water table, so that the total head is zero at all places. First we plot the solute concentration versus depth. For the clay we must assume some sort of "average" value to be representative of both the micro- and macroporosity, since we have already established that concentration varies locally within this material. Next, recognizing that the solute potential is approximately proportional but opposite in sign to the solute concentation, we sketch in the solute head and elevation head curves. From (68) and our requirement that the total head be zero at all places, we can construct the pressure head curve. The pore pressure is then given by $u = \gamma_w\ g\ h_p$ and, since it has the same shape as the pressure head curve, we see immediately that the pore pressures are not hydrostatic and are influenced by the osmotic coupling.

When we attempt to measure the head distribution using our (transducer-type) piezometer filled with pure water, we know from (75) that

$$h_{pmeas} = h_p + h_s \qquad \text{(clay)} \qquad (76)$$

for the clay stratum, but since the rock of the saline aquifer cannot act as an osmotic membrane between its own pore water and the piezometer water

$$h_{pmeas} = h_p \qquad \text{(aquifer)} \qquad (77)$$

The overall curve for h_{pmeas} is sketched in and reveals a normally pressured clay

underlain by an overpressured saline aquifer. *In the absence of information on pore water chemistry we would interpret this as representing a steep total head gradient at the lower aquifer / clay interface and we would infer an upward water flux where none exists.*

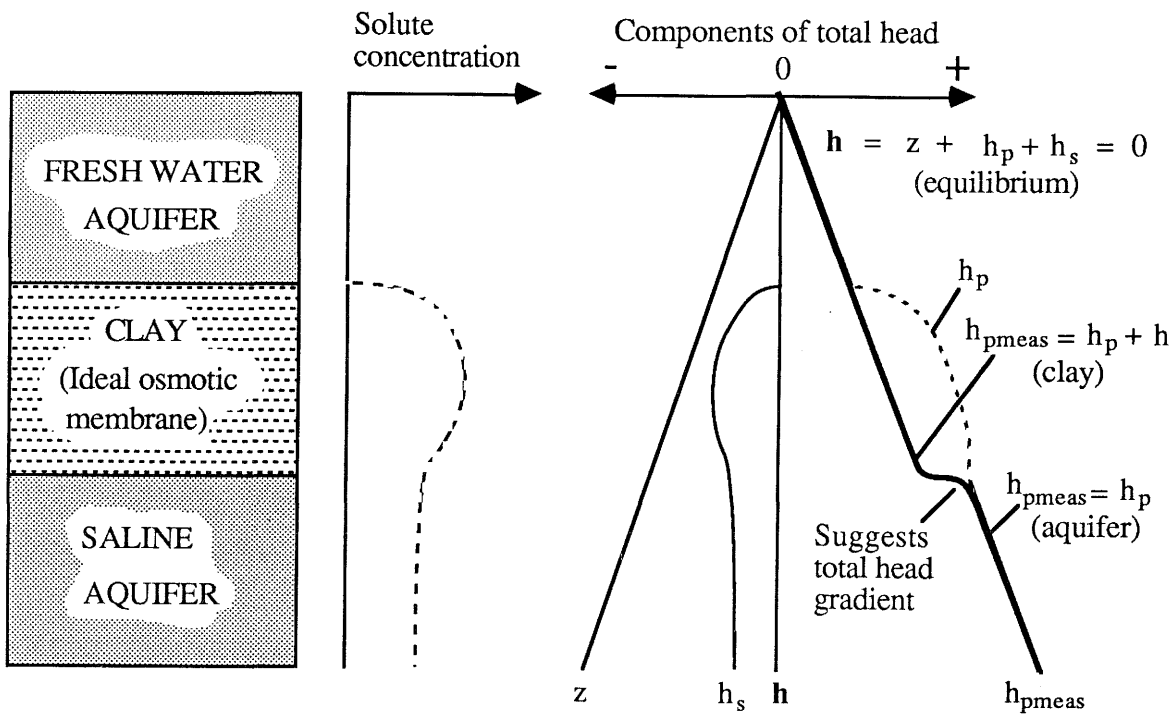


Fig. 13: Idealized situation of a clay membrane separating fresh water and saline aquifers. Zero vertical groundwater flux is assumed. The measured pressure heads h_{Pmeas} in this system would suggest that a total head gradient exists at the clay / saline aquifer interface. In the absence of information on solute chemistry we would incorrectly infer that a vertical (upward) flux exists across this interface.

One weakness in the above discussion is that we have ignored other possible physico-chemical reactions between water and clay, such as hydration which could have profound, but different, effects on pressure head measurements in highly-compacted clays and shales. We also ignore the practical difficulties of head measurement using a piezometer and assume an ideal, non-compliant instrument emplaced without disturbance to the clay. Nevertheless we feel that the issues raised in this section illustrate some of the difficulties in applying standard hydrogeological methods and concepts to clays.

6.4 Borehole Effects

It is not possible to introduce a hydrogeological measuring system into a clay stratum without disturbing the ground around the instrumentation. The act of drilling a borehole in a rock under stress results in considerable perturbation of the stress state in material adjacent to the hole. Any change in the total stresses acting on a porous medium will produce a finite and predictable change in pore pressure, and in a compressible medium such as clay, the induced pore pressure changes may be quite considerable. In addition, the borehole flushing fluid and the water introduced into the hydrogeological testing system will rarely be at formation temperature and are often substantially cooler. Cooling of the borehole walls and, subsequently, of the rock more remote from the hole will produce additional perturbation of the stress state and pore pressures. Finally, the introduction of fluids into the hole during drilling and testing, which have very different chemistries from those of the formation water, will produce swelling, oxidation and other chemical reactions. These will further complicate the situation.

This problem becomes even more complex when we examine the displacement of the borehole wall produced by stress and pore pressure changes in material adjacent to the hole. Some displacement will occur during the drilling phase and will not influence the measurements. However, additional movement will occur as the stresses and pore pressures slowly re-equilibriate, and as the formation imbibes water during swelling. Large displacements may also be engendered if the water in the test section is pumped down during the progress of a permeability measurement. The effect of borehole wall displacements on head measurements will depend very much on the design of the instrumentation. If the test section has low volume and is equipped with strain gauge type pressure transducers it will have low compliance. *In this case, inward motion of*

the borehole walls will produce significant pressure changes in the test section. In more compliant set-ups the effect will be less pronounced.

In permeable rocks, many of the borehole effects described above do occur, but their effect is to produce transients of such short time duration that equilibrium can readily be achieved during head measurements. Also, since the elastic moduli of most permeable rocks are moderately high and swelling is uncommon in these rocks, borehole closure can be ignored.

In low permeability clays, the duration of transients produced by the above effects is likely to be very long and extended time periods must elapse before the pressure indicated by the measuring system stabilises. If this was the whole story then we could simply resign ourselves to the fact that measurements in compacted clays take a long time and move ahead with the testing programme on that basis. However, can we be sure that the pressure response in clay is exactly analogous to that in a more permeable rock? If the response of the clay around the boreholes was exclusively due to pore pressure, stress and temperature re-equilibriation and the borehole wall displacements were generally small then we would have reasonable grounds for assuming this to be so. However, for clays under high shear stresses, the displacements are likely to be large and possibly time-dependent. Furthermore we have the very difficult problem of chemical interaction between the water in the test section and the clay, resulting in swelling and other effects.

In the last section we discussed the components of total potential for advective flow in clays and showed that, at equilibrium, our ideal (transducer-type) piezometer filled with pure water shows a measured pressure head at a particular elevation given by

$$h_{pmeas} = h_p + h_s \qquad (78)$$

where h_p is the pressure head at one particular point in the clay and h_s is the solute head at that same point. The reason for specifying a particular point was that the relative magnitudes of h_p and h_s may be different at another point in the clay with only the sum of these terms remaining constant (at one elevation). By insisting on the ideal, non-compliant piezometer we were able to avoid the issue of water mixing within the piezometer and, by assuming that the clay behaved as an ideal membrane, we avoided the issue of solutes diffusing into the test water. In practice mixing of the water in the

piezometer with that of the formation and the diffusion of solutes from the formation will occur, and the solute head in the piezometer water cannot be assumed to be zero at all times. Diffusion will only cease when the solute concentration gradients have been eliminated. *This particular form of transient could be of considerable duration.*

We are faced with the difficult task of trying to assess the magnitude of these processes and their probable influence on hydrogeological testing. We recognize, immediately that we are dealing with an extremely complicated coupled system, involving not only the mechanical, thermal, hydraulic and chemical responses of the rock but also those of the testing system. Although we feel that a model can be developed incorporating all these coupled processes, we confine ourselves for the time being to a simpler approach whereby certain of the more important processes are examined in isolation.

Answering the question raised above, hydrogeological tests in compacted clays are not analogous with those in more permeable media. *Differences are not simply the duration of the transients, but more fundamental ones associated with borehole effects, coupled flow and the complex interaction between the test fluid and the formation.*

As a final thought on this issue if, as we imply in Section 3.1, most of the water in intact Opalinus Clay is actually adsorbed on the surface of clay minerals, then hydrogeological tests in *unfractured test intervals* may reveal nothing whatsoever about the process of advection in this material. *The measured test response might simply be a consequence of the physico-chemical interaction between fresh water and highly compacted clay minerals.*

6.4.1 Swelling

Swelling in clays is the physico-chemical process by which free water is drawn into the material resulting in a volume increase. If the clay is constrained so that volume change is not possible, then a swelling pressure is developed which acts against the constraint.

Swelling can present major difficulties in borehole drilling in clay and shale formations. Penetration of the ground by the drill bit is accompanied by significant readjustment of the stresses and by swelling as the clay takes in water from the drilling mud. In strong formations, swelling pressure may increase the hoop (tangential) stress in the borehole wall resulting in "hydration spalling" and caving. Research relating to oilfield drilling

has shown that swelling pressures as large as 35 MPa may be developed in problem shales (KELLY, 1969). Swelling in such materials may be controlled, to some extent, by the use of specially formulated drilling muds with chemical additives (GRAY et al., 1980).

Similar difficulties are encountered in tunnelling where the swelling of clays and shales can produce substantial and problematic inward movements (heave) of the rock during excavation and can severely overload temporary support systems and permanent linings (EINSTEIN, 1979). A number of the marls and clay-shales of Switzerland, including the Opalinus Clay, have presented particularly severe tunnelling problems (GROB, 1976; KOVARI, MADSEN and AMSTAD, 1981; MADSEN and MULLER-VONMOOS, 1985).

We note that swelling is likely to have a finite but, as yet unquantified, effect on the *permeability* of a mudrock.

In spite of the considerable volume of research on the subject, *the precise mechanisms of swelling in clays and shales remain uncertain* (HUANG et al., 1986). We distinguish immediately between *intraparticle* swelling, or the introduction of intermicellar water into specific clay minerals such as smectite, and *interparticle* swelling which occurs in most natural clays regardless of their precise clay mineralogy. Current concepts of interparticle swelling postulate the existance of forces of attraction and repulsion between the clay platelets. The forces of attraction comprise the so-called van der Waals forces plus electrical forces between positively charged particle corners or edges which may be cross-linked to negatively charged particle faces. Both of these sets of forces act over very short distances. The van der Waals forces are largely independent of the fluid in the interparticle space, but the electrical forces of attraction are affected to a degree by the physical properties of the fluid. The forces of repulsion have two main components, first, forces of hydration at the surfaces of the particles and, second, osmotic forces produced in the diffuse double layers of the aqueous phase. The forces of hydration operate over distances of only a molecular order of magnitude, whereas the osmotic forces are effective over much greater distances of a maximum order of 0.1 μm (HARDY, 1965).

In compacted clays, interparticle forces associated with the elastic distortion of contacting mineral particles may contribute to the net repulsion tending to produce

swelling. In overconsolidated clays, interparticle or "contact" forces are the source of a considerable amount of recoverable strain energy (BJERRUM, 1967).

The repulsive forces between clay particles are potentially much greater than the attractive forces. In the field situation swelling is restricted by the stresses acting within the clay stratum and by the low permeability of the medium. When the total stress is removed and free water is made available, then volume changes are no longer constrained and the mechanisms of swelling become operative. Interparticle repulsion is manifested not only in swelling but also in the rebound which accompanies unloading in clays, and mechanistically swelling and rebound may be regarded as identical processes.

The development of diagenetic bonds and the introduction of a cementing medium may limit the capacity of a clay or shale to swell on the removal of stress. Weathering processes may however cause the physical breakdown of interparticle bonds and cement enabling swelling to occur over extended time periods. We note in this context that conditions of low pH resulting from the oxidation of pyrite may accelerate the weathering process and trigger swelling.

The relative significance of the hydration and osmotic components of interparticle repulsion is controversial (LOW, 1987). In highly compacted clays with, interparticle spacings which approach molecular dimensions, hydration may be the dominant mechanism of swelling. At clay particle spacings of less than 1.5 nm, the exchangeable ions are uniformly distributed in the interparticle space and do not separate into diffuse layers. Hydration of the clay mineral surfaces, or possibly of the ions within the interparticle space, would tend to force the clay particles apart. *Quantitative predictions of swelling by the hydration mechanism have yet to be made.*

At higher water contents the osmotic component of repulsion may assume greater significance. LANGMUIR (1938) assumed that the repulsive pressure tending to force particles apart equals the osmotic pressure midway between their parallel surfaces less the osmotic pressure of the external equilibrium solution. The osmotic pressure is then given by the van't Hoff equation

$$P = kT \sum (n_{im} - n_{i0}) = \mathbf{R}T \sum (C_{im} - C_{i0}) \tag{79}$$

where k is Boltzmann's constant, T is absolute temperature, **R** is the gas constant, n_i is the ionic concentration (ions.l^{-1}) and C_i is the molar concentration (mol.l^{-1}). The subscript m signifies the midplane between the surfaces of the particles, and the zero subscript signifies the external equilibrium solution.

For a single cation and anion species of the same valence

$$P = \mathbf{R}T\left[C_m^+ + C_m^- - C_0^+ - C_0^-\right] \tag{80}$$

where the + and - superscripts denote the cation and anion, respectively. It may be shown that, at equilibrium, in a dilute solution

$$C_0^+ = C_0^- \quad \text{and} \quad C_m^+ \cdot C_m^- = C_0^+ \cdot C_0^- = C_0^2 \tag{81}$$

where C_0 is the solute concentration of the external solution (mol.l^{-1}). Thus (80) becomes

$$P = \mathbf{R}T\left[C_m^+ + \frac{C_0^2}{C_m^+} - 2C_0\right] \tag{82}$$

If the cation concentration at the midpoint between particles is considerably greater than the solute concentration in the external solution then the second term within the parentheses on the right hand side of (82) is small and can be ignored giving

$$P = \mathbf{R}T\left[C_m^+ - 2C_0\right] \tag{83}$$

The distribution of ions as a function of distance from the clay surface can be calculated according to the Poisson-Boltzmann theory (GOUY, 1910; 1917; CHAPMAN, 1913).

According to double layer theory the ion concentration midway between the clay surfaces is determined by the value of the electrical potential ψ_m at this point, and it can be shown that

$$P = 2 n_0 k T \left[\cosh \frac{\nu e \psi_m}{k T} - 1 \right] \tag{84}$$

where n_0 is the ionic concentation in the external solution (ions.unit volume^{-1}), ν is the valence, and **e** is the elemental charge (VERWEY and OVERBEEK, 1948; BOLT, 1955; 1956; VAN OLPHEN, 1977).

With a number of additional assumptions, the osmotic repulsion P can be estimated from basic physical and mineralogical properties of the clay (MITCHELL, 1976; VAN OLPHEN, 1977; MADSEN and MULLER-VONMOOS, 1985). Important parameters are the half spacing between clay particles, which is related to water content, internal specific surface area and clay fraction, valence of the ions, cation exchange capacity (CEC) of the clay, concentration of the external solution, and temperature. Pressure P rises substantially with decreasing water content (Appendix C).

Conventionally, the total stress is carried by the effective stress and the *measurable* pore pressure (pore pressure determined in the normal way, using pure water in the test section)

$$\sigma = \sigma' + u_0 \tag{85}$$

If we make the assumption that the *repulsive force in a swelling clay is primarily due to double layer interactions, then the conventional (one-dimensional) effective stress* σ' *can be considered to be made up of three components*

$$\sigma' = \sigma_i' + R - A \tag{86}$$

The first component σ_i' is the interparticle stress which is the stress carried by particle to particle contacts within the material. The second component R is the (long-range) osmotically-derived repulsive force per unit area between clay platelets associated with

double layer interaction (the osmotic pressure is referenced to *pure water*). The third component A is the attractive force per unit area between clay platelets due to van der Waals and electrical forces (LAMBE, 1960). Substituting (86) into (85) we obtain

$$\sigma = \sigma_i' + R - A + u_0 \tag{87}$$

The repulsive force per unit area R is related to the solute head midway between the clay platelets by

$$R = - \gamma_w g h_{sm} \tag{88}$$

In Section 6.3 we showed that at equilibrium the pore pressure u_0 measured using a piezometer filled with pure water is given by

$$u_0 = \gamma_w g (h_{pp} + h_{sp}) = \gamma_w g (h_{pm} + h_{sm}) \tag{89}$$

where h_{pp} and h_{sp} are the pressure head and solute heads within a macropore and h_{pm} is the pressure head midway between two clay platelets. The hydrostatic pressure in the macropores u_p is therefore

$$u_p = \gamma_w g h_{pp} = u_0 - \gamma_w g h_{sp} \tag{90}$$

Substituting (88) and (89) into (87) and using (90) we obtain

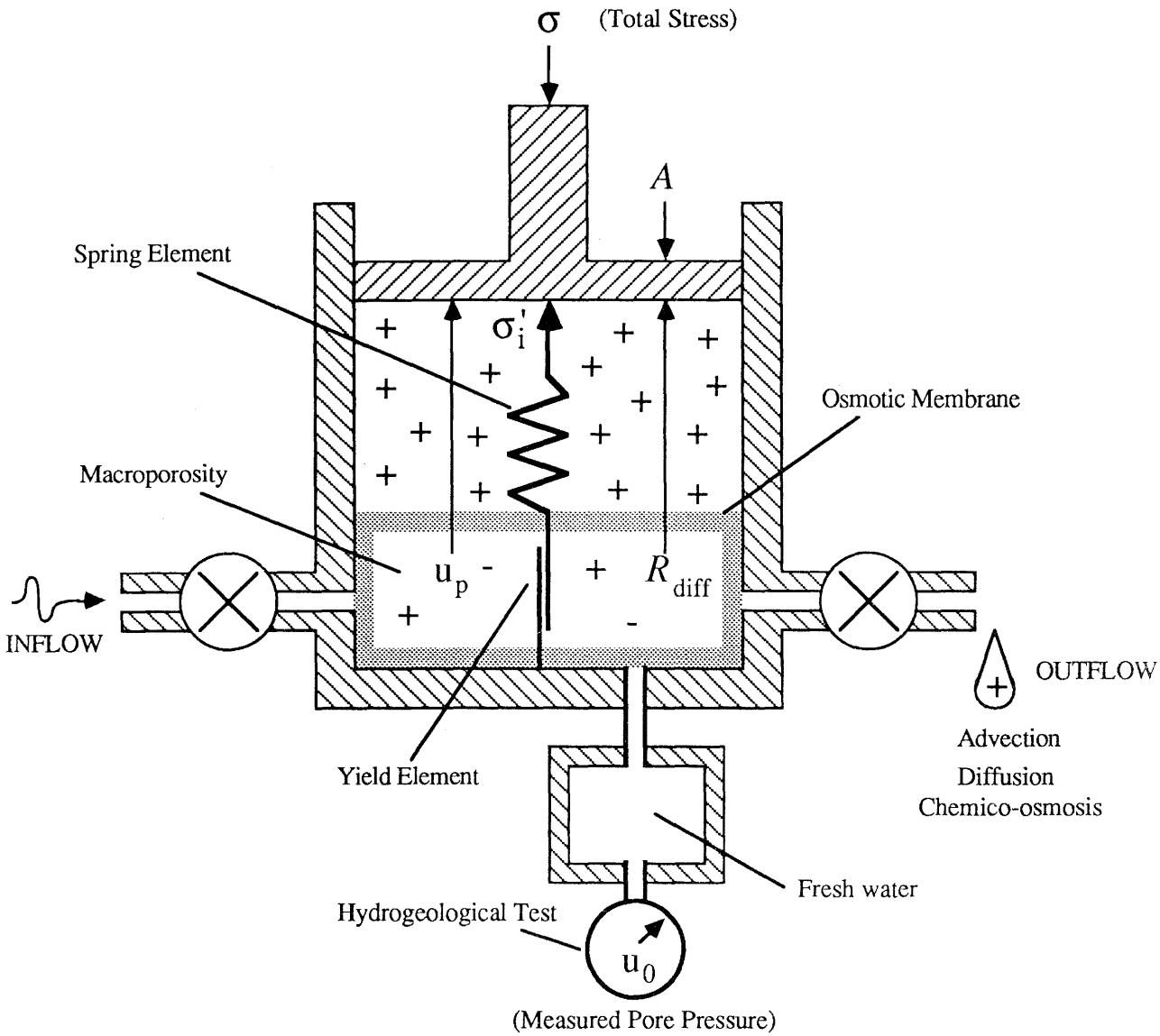
$$\sigma = \sigma_i' - \gamma_w g h_{sm} + \gamma_w g h_{sp} - A + u_p \tag{91}$$

If we define R_{diff} by

$$R_{diff} = - \gamma_w g \left[h_{sm} - h_{sp} \right] \tag{92}$$

we obtain

$$\sigma = \sigma_i' + R_{diff} - A + u_p \tag{93}$$



$$\sigma = \sigma'_i + R_{diff} - A + u_p$$

Fig. 14: Simple one-dimensional analogue for the hydro-chemico-mechanical responses of a swelling mudrock. The total stress σ on the mudrock is borne by the interparticle stress σ_i', the pore water pressure u_p, and the net physico-chemical forces per unit area acting between the clay platelets (R_{diff} - A). Pressure u_0 measured in a hydrogeological test is influenced by osmosis.

Fig. 14 shows a one-dimensional analogue for the hydro-chemico-mechanical responses of a swelling mudrock which illustrates these concepts. The analogue comprises a cylinder containing a moving piston. The total stress (due to the weight of overburden) acts downward on the piston. The reaction to the total stress is provided by the interparticle stress of the spring element σ_i', the pore pressure within the macropores u_p, and the net physico-chemical forces per unit area acting between the clay platelets (R_{diff} - A). The R_{diff} term is the (long-range) osmotically-derived repulsive force per unit area between clay platelets (due to the *concentration difference* between the water in the midplane region and the macropores). The yielding element in Fig. 14 is incorporated to enable the analogue to sustain large swelling strains upon reduction or removal of total stress ("free swell" conditions). The schematic "hydrogeological test" is included to illustrate the point that the measured pressure u_0 is *not equal* to the pore water pressure u_p in the macropores of the mudrock.

If we take C_0 in (83) to be the solute concentration within the macropores of the mudrock, then

$$R_{diff} = P = \mathbf{R}\,T\left[C_m^+ - 2\,C_0 \right] \tag{94}$$

and

$$u_p - u_0 = R - R_{diff} = 2\,\mathbf{R}\,T\,C_0 \tag{95}$$

6.4.1.1 Trial Calculations on Borehole RB26B

We are now in a position to perform some simple, order of magnitude, calculations on the effect of (osmotic) swelling on the RB26B borehole measurements. We will ignore borehole stress concentrations and the complications of a three-dimensional stress field.

Assuming a vertical total stress gradient G_S of 0.025 MPa.m^{-1}, the total stress σ at the depth of each of the four measuring points may be calculated. The water pressures u_0 in the gravel packs at the end of the initial period of head decline are then simply obtained from the total heads **h** (see Section 6.2).

We *assume* that the pressure head h_{pp} within the macropores of the clay is given by the height of a column of water extending upwards to the water table. Thus the pore water pressures u_p may be calculated for each measuring point (Table 5A).

Table 5A: Trial Calculations on Borehole RB26B

Pressure Transd.	Depth z (m.b.g.s)	Total Stress σ (MPa)	Total Head **h** (m)	Meas. Water Pressure u_0 (MPa)	Assumed PorePress. u_p (MPa)
PZ6	24.0	0.60	-24	0.00	0.23
PZ7	40.0	1.00	-40	0.00	0.39
PZ8	59.8	1.50	-56	0.04	0.58
PZ5	80.0	2.00	-76	0.04	0.77

Table 5B: Trial Calculations on Borehole RB26B (cont.)

Pressure Transd.	u_p-u_0= R-R_{diff} (MPa)	Solute Conc. C_0 (mol.l^{-1})	σ-u_p= σ_i'+R_{diff}-A (MPa)	R_{diff} (MPa)	R (MPa)
PZ6	0.23	0.049	0.37	0.19	0.60
PZ7	0.39	0.081	0.61	0.31	0.70
PZ8	0.54	0.112	0.92	0.46	1.00
PZ5	0.73	0.152	1.23	0.62	1.35

Using (95) we can now estimate R - R_{diff} and the solute concentration C_0 (mol.l^{-1} NaCl) of the water in the macropores (Table 5B). The analysis suggests that C_0 increases with depth from 0.05 mol.l^{-1} at 24 m to 0.15 mol.l^{-1} at 80 m. The predicted range of C_0 is not unreasonable for near surface Opalinus Clay (see experimental results discussed in Section 2).

Having evaluated σ and u_p, we can now use (93) to calculate $\sigma_i' + R_{diff} - A$.

Double layer theory, presented in Appendix C, provides a way of estimating R_{diff} directly from the physical and mineralogical properties of the clay. The mean water content of RB26B core is 5.2% (equivalent to our generic "shallow" Opalinus Clay of Section 4.2). Using the method outlined in Section 3.1, we estimate the mean half distance d between clay platelets to be circa 0.57 nm. Taking the valence of cations within the double layer as 1, the specific surface of the clay fraction S as 140 m^2.g^{-1}, the cation exchange capacity of the clay as 25 meq.100g^{-1}, temperature T as 293°K and taking the solute concentration of the (external) macropore water to be in the range 0.05 - 0.15 mol.l^{-1} NaCl, suggests R_{diff} to lie in the range 2.8 - 3.3 MPa. We must acknowledge, however, that these calculations for R_{diff} are subject to *very great uncertainty.*

In view of the uncertainty in evaluating from R_{diff} from first principles, we choose a different approach in order to pursue our scoping calculations. We make the *arbitrary assumption* that R_{diff} is equal to one-half of the calculated quantity $\sigma_i' + R_{diff} - A$, or

$$R_{diff} = \frac{1}{2}(\sigma_i' + R_{diff} - A) \qquad (96)$$

recognising that the values generated in this way are somewhat lower than those obtained by double layer calculations. Equation (95) then allows us to estimate R. Values are presented in Table 5B.

Laboratory measurements of the swelling pressure developed by the Opalinus Clay when exposed to pure (distilled) water provide additional evidence which is generally supportive of this interpretation. The one-dimensional swelling pressure test is performed on a laterally-constrained undisturbed sample and determines the maximum stress developed when distilled water is admitted to the sample and volume change is

prevented (see, for example, HOBBS et al., 1982). Although the relationship between swelling pressure and the various interparticle stress components has yet to be fully elucidated, it is often assumed that swelling pressure is numerically identical to R, the long-range, osmotically-derived, repulsive stress (referenced to pure water).

MADSEN and MULLER-VONMOOS (1985) tested core samples of Opalinus Clay from exploratory boreholes drilled at the Brugg Tunnel site in Northern Switzerland. One-dimensional swelling pressures for this material were in the range 0.7 - 2.2 MPa, with a mean of 1.3 MPa (19 tests). Our "first order" estimates for R given in Table 5B (0.60 - 1.35 MPa) are evidently in good agreement with the laboratory data.

Our calculations are too uncertain to make any definitive statements about the source of the anomalous heads in RB26B. However, the observations are *consistent with the hypothesis that the total head* **h** *in a mudrock has three components*

$$\mathbf{h} = z + h_{pp} + h_{sp} \tag{97}$$

where z is the elevation head, and h_{pp} and h_{sp} are the macropore pressure and solute heads, respectively. The pressure u_0 registered by a transducer installed in a test zone containing fresh water is then

$$u_0 = \gamma_w g \left[h_{pp} + h_{sp} \right] = u_p - 2\,\mathbf{R}\mathbf{T}\,C_0 \tag{98}$$

where u_p is the water pressure in the macropores and C_0 is the (monovalent) solute concentration of the porewater. If $C_0 > 0$ then $u_0 < u_p$ and the *observed head will be anomalous.*

Another interesting relationship is

$$u_p - u_0 = R - R_{diff} \tag{99}$$

where R is the long-range, osmotically-derived, repulsive force per unit area between clay platelets referenced to *pure water*, and R_{diff} is an equivalent osmotic force per unit area referenced to the *macropore water solute concentration.* At least as an approximation, R may be equated with the laboratory-determined swelling pressure.

We note that the long-term upward drift of the head measurements, after the initial period of head decline, might be explained by solutes diffusing from the mudrock to the test water in the gravel packs.

The most important conclusion that we draw from this section is that *chemico-osmotic (coupled) flow and the physico-chemical process of swelling in overconsolidated mudrocks are closely related.* It is probably no coincidence that the characteristics of a clay which render it an efficient osmotic membrane are identical to those which endow it with a high capacity to swell.

6.4.2 Borehole Stresses (Undrained Analysis)

In this section we briefly examine the stresses and induced pore pressures around a vertical borehole drilled in a mudrock formation. The analysis assumes simple elasto-plastic behaviour and no advective flow during deformation (i.e. an undrained analysis). The solutions are therefore applicable to the period shortly after drilling. Osmotic effects on pore pressure are ignored.

A vertical cylindrical opening is examined under plane strain conditions (vertical strain $\varepsilon_z = 0$). We assume Poisson's ratio to be 0.5 in both the elastic ("quasi-elastic") and plastic states. The horizontal total stresses are assumed to be equal to the overburden stress σ_v. Plastic yielding is assumed to occur when the shear stresses reach the undrained shear strength S_u (= half uniaxial compressive strength) of the mudrock. The angle of internal friction for undrained loading is taken as zero.

Based on SERATA and GLOYNA (1960), FARA and WRIGHT (1963) and DEERE et al. (1969), the radius of the plastic zone R around a borehole is given by

$$R = a \exp\left[\frac{\sigma_v - S_u - P_{bore}}{2 S_u}\right] \qquad (100)$$

where a is the borehole radius and P_{bore} is the fluid pressure within the borehole. Thus plastic yielding occurs when

$$\sigma_v > (S_u + P_{bore}) \tag{101}$$

The radial, tangential and vertical total stresses in the plastic zone ($a \leq r \leq R$) are given by

$$\sigma_r = 2\, S_u \ln\left[\frac{r}{a}\right] + P_{bore} \tag{102}$$

$$\sigma_t = 2 \cdot S_u \cdot \left[1 + \ln\left[\frac{r}{a}\right]\right] + P_{bore} \tag{103}$$

$$\sigma_z = S_u \cdot \left[1 + 2 \ln\left[\frac{r}{a}\right]\right] + P_{bore} \tag{104}$$

where r is the radial distance from the centreline of the hole. The total stresses in the elastic zone ($r > R$) are given by

$$\sigma_r = \sigma_v - S_u \left[\frac{a}{r}\right]^2 \exp\left[\frac{\sigma_v - S_u - P_{bore}}{S_u}\right] \tag{105}$$

$$\sigma_t = \sigma_v + S_u \left[\frac{a}{r}\right]^2 \exp\left[\frac{\sigma_v - S_u - P_{bore}}{S_u}\right] \tag{106}$$

$$\sigma_z = \sigma_v \tag{107}$$

The stress changes produced by the drilling of the borehole have a significant effect on the pore pressures in the mudrock adjacent to the hole. As a first approximation, the pore pressure increments Δu at each point in the rock can be calculated from the stress increments

$$\Delta u = \frac{B}{3}\left[\Delta\sigma_r + \Delta\sigma_t + \Delta\sigma_z\right] + A\left[(\Delta\sigma_r - \Delta\sigma_t)^2 + (\Delta\sigma_t - \Delta\sigma_z)^2 + (\Delta\sigma_z - \Delta\sigma_r)^2\right]^{\frac{1}{2}} \tag{108}$$

where A and B are Henkel's modified pore pressure parameters. Pore pressure parameter B simply describes the effect of changing mean normal total stress and, as we showed in Section 4.4 , B for a saturated rock is related to the Biot poroelastic constants R and H by

$$B = \frac{R}{H} \tag{109}$$

which we showed to be sensibly close to unity for the Opalinus Clay. Pore pressure parameter A describes the effect of octahedral shear stress changes on pore pressure. In a given soil or rock, A varies as shear stress increases towards failure. In a heavily overconsolidated mudrock, A at failure can be negative, signifying a tendency to dilate.

By way of illustration, Fig. 15 shows the total stress and pore pressure distributions at a point 400 m below surface around a borehole in the Opalinus Clay. We assume a uniaxial compressive strength of 10 MPa which gives S_u = 5 MPa. The vertical stress gradient is taken as 0.0226 $MPa.m^{-1}$ (1 $psi.ft^{-1}$) and the pore pressures prior to drilling are assumed hydrostatic. The total head of the borehole fluid is taken as -300 m with respect to a surface datum and the density of water in the borehole and the formation is taken as 1.0 $Mg.m^{-3}$. Additionally, we assume that A = -0.2 in both the plastic and the "quasi-elastic" states and that B = 1. We note in this context that no closed-form solutions are available which allow A to be treated as a stress-dependent variable.

The effect of stress change on pore pressure is most apparent in the plastic zone close to the borehole. The pore pressures in this region are slightly negative signifying a pore water suction. Thus there is a *tendency for the mudrock to draw water into this area from the borehole and from the surrounding formation.* Furthermore, movement of water from the borehole to the formation will occur whenever the borehole water pressure exceeds formation pore pressure. We emphasise that this mechanical effect is *additional* to the chemico-osmotic process of swelling discussed in the last section.

Since the pore pressure distribution predicted by this analysis differs from that assumed in the hydrogeological interpretation of permeability tests, *data reduction formulae derived for stronger and more permeable media may be inappropriate for clays.*

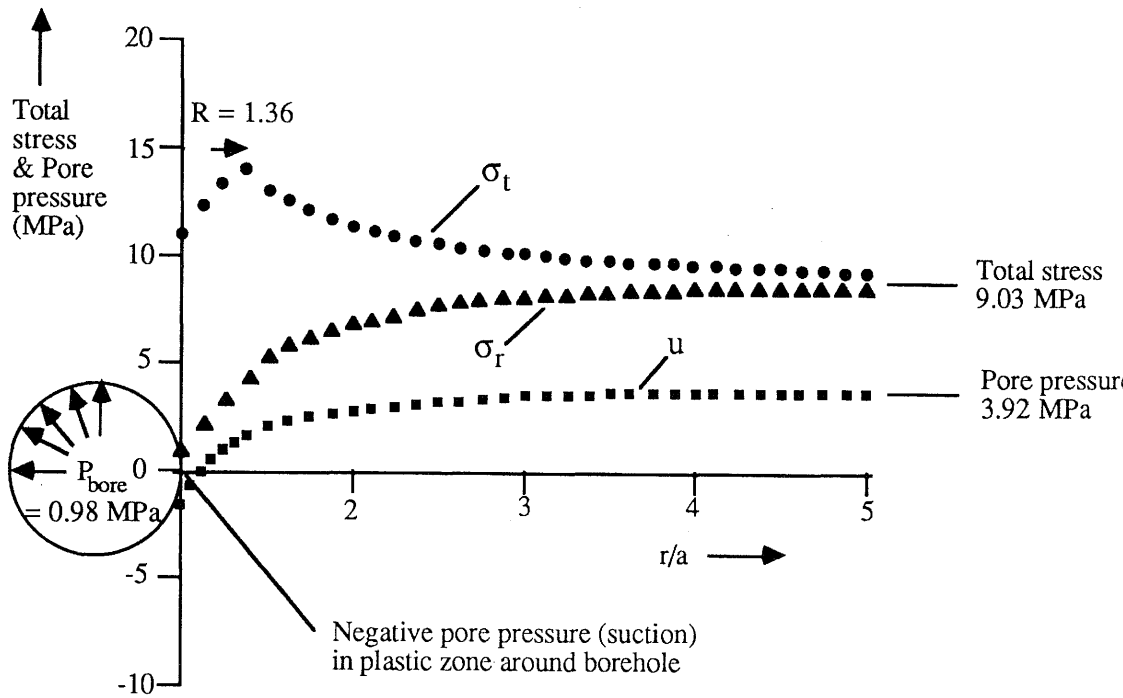


Fig. 15: Total stress and pore pressure distribution around a borehole in the Opalinus Clay based on a simple elasto-plastic model. A depth of 400 m is assumed and the undrained shear strength of the clay-shale is taken as 5 MPa.

If we examine the specific case of borehole RB26B at the site of the proposed Wisenberg Tunnel, then, at 80 m depth, with σ_v = 2 MPa, P_{bore} = 0.04 MPa and S_u = 5 MPa, we find

$$\sigma_v < (P_{bore} + S_u) \qquad (110)$$

which suggests that plastic yielding will not occur. However, we must recognise the possibility that swelling pressures, developed as the clay imbibes water, may be of sufficient magnitude to produce localized yielding around the borehole. Nevertheless, we feel that stress-related borehole effects are likely be comparitively small in these shallow boreholes.

7 LIMITATIONS OF THE "SOLUTE POTENTIAL" CONCEPT

In Section 3.2 we introduced the soil mechanics concept of "solute potential" ϕ_s as a component of the total potential associated with water flow in a mudrock

$$\phi = \phi_z + \phi_p + \phi_s \qquad (111)$$

In subsequent chapters we made considerable use of the analogous "head components" relationship

$$\mathbf{h} = z + h_p + h_s \qquad (112)$$

in our examination of osmotic phenomena (membrane effects and swelling). In this chapter we shall demonstrate that the terms "solute potential" and "solute head" can only be rigorously applied in very simple idealized systems.

Consider the situation shown schematically in Fig. 16. An aquifer, exposed at surface (left hand side of diagram), dips down below a mudrock stratum which acts as an ideal osmotic membrane (right hand side of diagram). The salinity of the aquifer increases with depth. The mudrock is overlain by a freshwater aquifer. Zero groundwater flux is *assumed* throughout the system. On the right hand side of the system we can sketch in elevation, pressure and solute head profiles which meet the requirement that the total head **h** is non-varying with depth (vertical head gradient zero). On the left hand side we can sketch in the three components of total head (ignoring density effects on h_p) and, after summation, we find that **h** becomes increasingly negative with depth. This of course implies the existence of a total head gradient within the fluid column on the left hand side of the diagram which, in turn, suggests a downward *advective* flux where none exists.

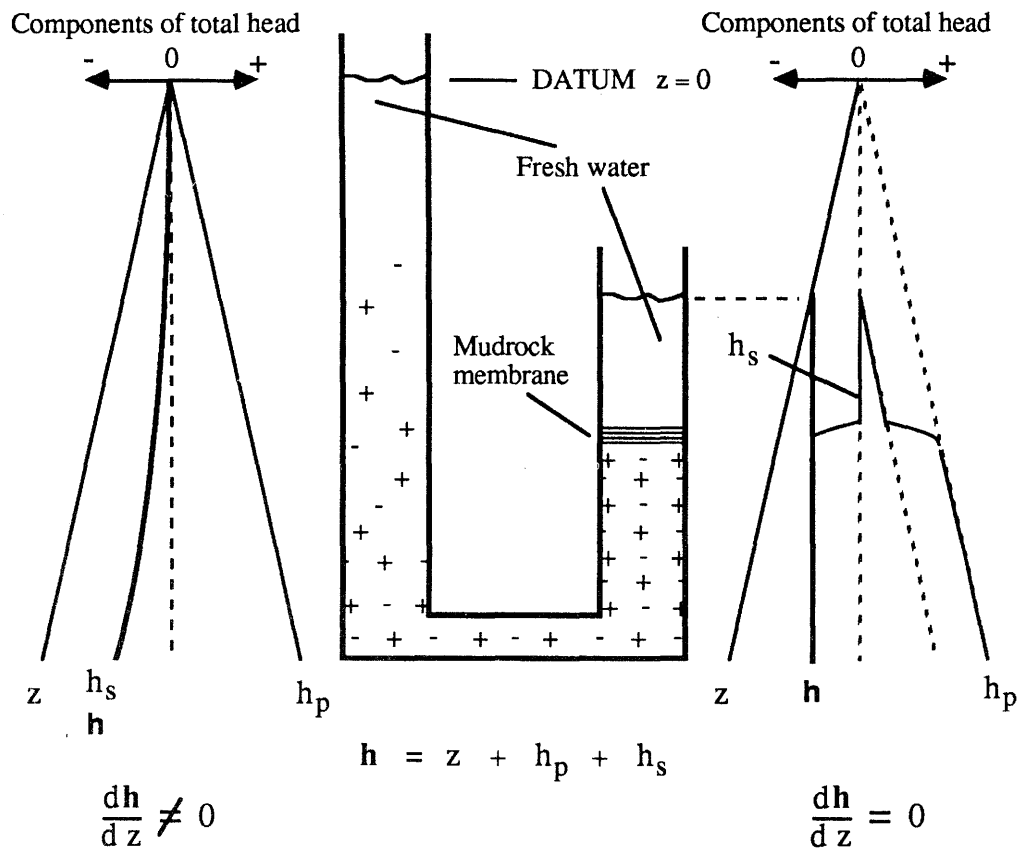


Fig. 16: Diagram illustrating the limitations of the solute head concept. An aquifer, exposed at the surface (left hand side of diagram), dips down below a mudrock stratum which acts as an ideal osmotic membrane (right hand side of diagram). The salinity of the aquifer increases with depth. The mudrock is overlain by a freshwater aquifer. Zero groundwater flux is assumed throughout the system. The head gradient inferred for the left hand side is incompatible with the assumtion of zero flux showing that solute head, as a parameter, is of limited value in the quantitative analysis of regional groundwater movement.

Our real difficulty lies in attempting to describe the combined effects of advection, diffusion and chemico-osmosis on groundwater movement using a single term, the total head **h**. The solute head component h_s of total head can only be usefully defined if the flow path under consideration *intersects or lies wholly within a mudrock stratum behaving as a ideal osmotic membrane*. Solute head cannot be usefully defined where concentration gradients exist in non-membrane materials or where membranes of varying degrees of ideality are present.

This three-fold subdivision of total head is therefore a considerable aid to conceptual thinking but has limited application in quantitative analysis.

8 CONCLUSIONS

What emerges from this preliminary study is that, although the mechanisms of groundwater movement in mudrocks are at present uncertain, certain processes could have such profound hydrogeological effects that their significance goes way beyond the interpretation of borehole test results, with repercussions in the *development of a strategy for site characterisation, in modelling and in safety assessment.*

Our calculations suggest that long-term transient flow may be occurring in the Opalinus Clay as a consequence of the stress changes associated with the process of overconsolidation and the removal of sedimentary cover by erosion. In addition, based on the strong evidence for neotectonic deformation of the crust in many areas of Switzerland, anomalous hydrogeological conditions may develop in low permeability rocks such as the Opalinus Clay as a result of stress- or strain-induced pore pressure changes.

The general picture which emerges from our calculations on the overconsolidation effect is one in which fracture flow fairly rapidly re-equilibriates after exhumation but re-adjustments of the water content of the intact mudrock may occur over a much longer time period, constrained somewhat by diagenetic bonding of clay minerals and by the presence of cements. Thus fracture flow might be regarded as being essentially out of phase with flow in the intact clay (matrix flow).

The low porosity and generally high clay content of the Opalinus Clay suggest that it might act as an efficient semi-permeable membrane supporting osmotically-driven flow not only across the stratum but also within it. Our calculations show that osmosis can have large effects on hydraulic head measurements and these must be considered when testing in mudrocks.

If we acknowledge osmotic flow to be a significant component of total flow in mudrocks, then long-term transient behaviour must be viewed as a response to hydraulic and chemical disequilibria.

Borehole effects associated with the mechanical, thermal and chemical changes occurring in the formation during drilling and testing can have a significant influence on the measured hydraulic response. Although swelling due to the introduction of fresh water is probably the most significant effect, in deep boreholes perturbation of the *in situ* stress field and plastic deformation of the rock may have a considerable effect on the pore pressure distribution around the borehole.

If we examine the particular case of the RB26B borehole, then the anomalously low heads can be explained (semi-quantitatively) by assuming that the total head **h** at each point in the hole is given by

$$\mathbf{h} = z + h_{pp} + h_{sp} \qquad (112)$$

where z is the elevation head, h_{pp} is the pressure head in the macropores, and h_{sp} is the solute head in the macropores. If this scenario is correct then it has very important implication to the design and interpretation of hydraulic tests in mudrock formations. It suggests that the hydraulic head determined in such tests is likely to be sensitive to the solute chemistry of the test fluid. It calls into question the common practice of using fresh water as the test fluid. Non-polar, non-reactive liquids might be more suitable for the purpose. Alternatively, the chemistry of the test fluid might be regarded as a variable in the development of a testing methodology for mudrocks.

9 RECOMMENDATIONS FOR ADDITIONAL WORK

We have highlighted a large number of uncertainties in our current understanding of groundwater flow in mudrock environments. Concepts and assumptions which are basic to the quantitative treatment of groundwater movement in more permeable rock-types become highly questionable when applied to low permeability mudrocks. At a fundamental level, the basic mechanisms of water movement in clays have yet to be properly identified and characterized, and we must acknowledge that mechanisms other than hydraulic (Darcy) flow probably play a significant role. In groundwater modelling, the assumption of normal gravitational flow under steady-state flow conditions does not bear close inspection, and we are forced to examine the hydrogeological implications of a number of long-term geological processes such as burial, exhumation and tectonic deformation. Finally, standard field methods for characterizing the hydrogeology of a site, including the measurement of heads and permeabilities in boreholes, are problematic when applied to mudrocks.

There is a pressing need to develop a testing methodology specific to these materials and to establish a theoretical framework for the interpretation of the test data.

The level of effort required to resolve these uncertainties and to furnish the necessary methodology and techniques for site characterization and safety assessment in a mudrock environment is very substantial. In the longer-term, this effort might include the development of a 2-D or 3-D regional groundwater / solute transport model, with transient flow capability, incorporating advection, diffusion and coupled flow. We recognize that the calibration of such a model would be a very demanding exercise requiring input from very carefully designed field experiments and specialised laboratory testing.

For the immediate future, we define three priority areas: (a) Sensitivity analysis of transient flow under hydraulic and chemical gradients, (b) Detailed examination of the hydrogeological implications of tectonic deformation, and (c) Development of a borehole testing methodology for mudrocks, together with the necessary theoretical models for reduction of the test data.

9.1 Sensitivity Analysis of Transient Flow under Hydraulic and Chemical Gradients

This activity would involve the development of a numerical model for 1-D transient flow under hydraulic and chemical gradients. The simultaneous differential equations describing advection, diffusion and osmostically-driven (coupled) flow would be solved for prescribed, time-varying, boundary conditions using the finite difference method. The effect of changing total stress would be considered. The model would accommodate horizontal layering (stratification) with vertical variation in material properties.

Geometries would range in complexity from a simple three-layer problem to a multilayer representation of a selected stratigraphical sequence (to include the Opalinus Clay). Provisional values for the material constants of the model would be established from a search of the literature and from known interrelationships. We also suggest a laboratory component to the study to provide data specific to the Opalinus Clay.

Stresses, pore pressures and pore wate solute concentrations would be specified at the boundary nodes. Specific geological processes, such as burial and exhumation, would be simulated by varying the boundary conditions with time in a prescribed manner.

The primary objectives of the study would be to determine the sensitivity of the predicted responses to the numerical values of the input parameters (as a guide to possible future data acquistion activities), and to obtain a better understanding of the overall performance of these systems.

We also note that laboratory experiments on coupled flow would most likely involve 1-D flow in laterally-constrained samples. Thus a secondary objective of the study would be to model this experimental geometry.

9.2 Hydrogeological Implications of Tectonic Deformation

We recognise that neotectonic deformation of the Opalinus Clay could play a dominant role in determining the hydrogeology of this formation. An important factor is the very strong coupling between the deformation and failure of the sediment and the pressure of the pore fluid. Both the volumetric and the shear components of strain can produce

changes in pore pressure in a deforming mudrock. If the rock mass is subject to inhomogeneous deformation, then we would anticipate the development of pore pressure gradients. The exchange of water between fractures and intact clay is also likely to be important mechanism.

As we noted in Section 4.2, quantitative analysis of this problem would require some development of the theoretical framework. The basic requirement is for a constitutive model that can describe both dilatancy and compaction of the rock under combined (3-D) states of stress. We suggest that an elasto-plastic, strain-hardening model based on "Critical State Concepts" could be usefully applied to this problem.

The study would explore the main characteristics of tectonically-induced groundwater movements in a deforming mudrock. Special attention would be paid to the role of faulting.

9.3 Borehole Testing Methodology for Mudrocks

We regard the development of a borehole testing methology specific to mudrocks, together with the necessary theoretical models for reduction of test data, as a matter of the highest priority. The case is made in this report that hydraulic tests in compacted clays and shales are not analogous with those in more permeable media. Differences are not simply the duration of the flow transients, but more fundamental ones associated with borehole effects, coupled flow and the complex interaction between the test fluid and the formation.

We should not underestimate the difficulties which are likely to be encountered in meeting our objective. One obvious constraint is our incomplete knowledge of the actual mechanisms of fluid flow in mudrocks. In order to make progress, we must raise the general level of understanding in this area and it is inevitable that our activities must include a significant component of fundamental research.

A key activity would be the development of numerical models capable of simulating the pressure and flow response during borehole testing. At the first level of sophistication the model(s) would have the following features: (a) Axisymetric, 1-D finite difference solution, (b) Transversely isotropic stress field, (c) Elasto-plastic constitutive law with dilatancy and compaction (consolidation), (d) Advection, diffusion and osmotically-

driven (coupled) flow, and (e) Swelling and borehole closure effects included. Thermal effects, ion exchange, adsorbtion and other features might be incorporated in more sophisticated versions.

The development of the modelling capability would be fully integrated with a programme of field experiments. These borehole experiments would be conducted in the Opalinus Clay and would be designed to test and refine the numerical models. An important conclusion of our report is that the hydraulic heads monitored *in situ* should be sensitive to the chemistry of the test fluid. Test fluid chemistry would be a variable in our "conceptual experiments". A broad range of laboratory tests would be performed on core samples, the more important parameters being pore water chemistry, clay mineralogy, swelling properties, strength and stress-strain properties and the transport properties (including coupled flow coefficients).

We feel that a study conducted broadly along these lines would not only provide a rationale for *in situ* testing in the Opalinus Clay, but would also provide a very valuable insight into the basic mechanisms of groundwater movement and solute transport in this formation. This would be invaluable in the preparation of a safety case.

ACKNOWLEDGEMENTS

This report was prepared as part of the NAGRA General Advisory Contract "Hydrogeological, Geochemical and Geomicrobiological Studies of the Disposal of High and Intermediate Level Radioactive Waste", PA83PD.

The writers wish to thank their colleagues M.A. Brightman and C.A.M. Ross of the Fluid Processes Research Group, BGS, for internally reviewing the manuscript and for their perceptive comments on a number of technical issues.

The report was externally reviewed by Dr C.J. Carnahan of Lawrence Berkeley Laboratory, University of California, and Drs F.T. Madsen, K. Kovari and G. Anagnoston of the Institute for Soil and Rock Mechanics, ETH-Zurich, and the constructive comments and suggestions of these individuals have been accommodated in the preparation of the finalised manuscript.

REFERENCES

ALEXANDER, J. (1990): A review of osmotic processes in sedimentary basins. British Geological Survey, Fluid Processes Research Group, Technical Rept. WE/90/12.

ALEXANDER, J. and HOLMES, D.C. (1983): The local groundwater regime at the Harwell research site. Rept. Institute of Geological Sciences (NERC), FLPU 83-1.

ANDERSSON, D.M. and LOW, P.F. (1958): The density of water adsorbed by lithium-, sodium-, and potassium-bentonite. Soil Science Society Proceedings, 99-103.

BARKER, C. (1972): Aquathermal pressuring: role of temperature in development of abnormal pressure zone. Am. Assoc. Pet. Geol. Bull., 56.

BEAR, J. (1972): Dynamics of fluids in porous media. Elsevier, New York.

BELITZ, K. (1985): Hydrodynamics of the Denver Basin: An explanation of subnormal fluid pressures. Ph D thesis, Stanford University, U.S.A.

BERRY, F.A.F. (1969): Relative factors influencing membrane filtration. Chemical Geology, 4, 295-301.

BERRY, F.A.F. (1973): High fluid potentials in California Coast ranges and their tectonic significance. Bull. Am. Assoc. Pet. Geol., 57, 1219-1249.

BERRY, F.A.F. and HANSHAW, B. B. (1960): Geologic field evidence suggesting membrane properties of shales (abstract). International Geological Congress, Copenhagen, Report Session, Norden. 21, 209.

BIOT, M.A. (1941): General theory of three-dimensional consolidation. J. Appl. Phys., 26, 182-185.

BIOT, M.A. (1955): Theory of elasticity and consolidation for a porous anisotropic solid. J. Appl. Phys., 26, 182-185.

BJERRUM, L. (1967): Progressive failure of slopes of overconsolidated plastic clay and clay-shale. J. Soil Mech. Found. Div., A.S.C.E., 93(1), 1-49.

BLAESI, H.R. (1987): Lithostratigraphie und Korrelation der Doggersedimente in den Bohrungen Weiach, Riniken und Schafisheim. Eclogae geol. Helv. 80(2), 415-430.

BOLT, G.H. (1955): Analysis of the validity of the Gouy-Chapman theory of the electrical double layer. J. Colloid Sci., 10, 206-219.

BOLT, G.H. (1956): Physico-chemical analysis of the compressibility of pure clays. Geotechnique, 6, 86-93.

BOLT, G.H. and MILLER, R.D. (1958): Calculation of total and component potentials for water in soil. Trans. Am. Geophys. Union, 39(5).

BORGESSON, L., HOKMARK, H. and KARNLAND, O. (1988): Rheological properties of sodium smectite clay. Technical Rept. 88-30, Svensk Karnbranslehantering AB (SKB), Stockholm.

BREDERHOEFT, J.D., BLYTH, C.R., WHITE, W.A. and MAXEY, G.B. (1963): Possible mechanism for concentration of brines in subsurface formations. Bulletin American Association Petroleum Geology, 42(2), 257-269.

BREDERHOEFT, J.D. and HANSHAW, B.B. (1968): On the maintenance of anomalous fluid pressures, I. Thick sedimentary sequences. Geol. Soc. Am. Bull., 79, 1097-1106.

BREDERHOEFT, J.D. and PAPADOPULOS, S.S. (1980): A method for determining the hydraulic properties of tight formations. Water resources Res., 16(1), 233-238.

BRIGHTMAN, M. ALEXANDER,J., and GOSTELOW, T.P. (1987): Groundwater movement through mudrocks: Measurement and Interpretation. Report British Geological Survey, FLPU 87-1.

BRONDERS, J. (1989): *In situ* determination of the macropermeability of a clay formation in view of assessing leakage and mass transfer in a deep argillaceous formation. *In:* Geological Disposal of Radioactive Waste: Annual Report for 1989, Commission of the European Communities, Brussels, RWD 90/1, 246-251.

BROWN, R.S.J. and KORRINGA, J. (1975): On the dependence of the elastic properties of a porous rock on the compressibility of the pore fluid. Geophysics, 40, 608-616.

BUCHER, F. (1975): Die Rechterscherfestigkeit natuerlichter Boeden, ihre Einflussgroessen und Beziehungen als Ergebnis experimenteller Untersuchungen. Mitteilungen des Institutes fuer Grundbau und Bodenmechanik Nr. 103, Eidgenoess. Techn. Hochschule Zuerich.

BURST, J.F. (1969): Diagenesis of Gulf Coast clayey sediments and its possible relation to petroleum migration. Bull. Am. Assoc. Pet. Geol., 53, 73-93.

CARNAHAN, C.L. (1985): Thermodynamically coupled mass transport processes in a saturated clay. Materials Research Society Proc., 44, 491-498.

CARNAHAN, C.L. (1989): Theory and calculation of water distribution in bentonite in a thermal field. Materials Research Society Symp. Proc., 127, 729-734.

CHAPMAN, D.L. (1913): A contribution to the theory of electrocapillarity. Phil. Mag., 25, 475-481.

COLTEN-BRADLEY, V.A. (1987): Role of pressure in smectite dehydration - effects on geopressue and smectite-to-illite transformation. Bull. Am. Assoc. Pet. Geol., 71(11), 1414-1427.

CORAPCIOGLU, M.Y. and BEAR, J. (1984): Land subsidence - a regional mathematical model for land subsidence due to pumping. *In* : Fundamentals of transport phenomena in porous media, Edited by J. Bear and M.Y. Corapcioglu, Proc. NATO Advanced Study Institute on Mechanics of Fluids in Porous Media, Delaware, USA.

DEERE, D.U., PECK, R.B., MONSEES, J.E. and SCMIDT, B. (1969): Design of tunnel liners and support systems. Rept. to US Dept. Transportation, OHSGT, Contract No. 3-0152 (NTIS No. PB 183 799).

DE SITTER, L.U. (1947): Diagenesis of oil-field brines. Bulletin American Association Petroleum Geology, 31, 2030-2040.

DICKEY, P.A. and COX, W.C. (1977): Oil and gas in reservoirs with subnormal pressures. Am. Assoc. Pet. Geol. Bull., 61(12) 2134-2142.

DIEBOLD, P. (1986): Erdwissenschaftliche Untersuchungen der Nagra in der Nordschweiz; Strömungsverhältnisse und Beschaffenheit der Tiefengrundwasser. Mitt. aarg. natf. Ges., 31, 11-52.

DIEBOLD, P. and MULLER, W.H. (1985): Szenarien der geologischen Langzeitsicherheit: Risikoanalyse für ein Endlager für hochaktive Abfälle in der Nordschweiz. Technical Report NTB 84-26, Nationale Genossenschaft für die Lagerung radioactiver Abfälle (NAGRA), Baden, Switzerland.

EINSTEIN, H.H. (1979): Tunnelling in swelling rock. Underground Space, 4, 51-61.

FARA, H.D. and WRIGHT, F.D. (1963): Plastic and elastic stresses around a circular shaft in a hydrostatic stress field, Trans. A.I.M.E., 226, 319-320.

FORSYTHE, G.E. and WASOW, W.R. (1960): Finite difference methods for partial differential equations, Wiley, New York.

FRITZ, S.J. (1986): Ideality of clay membranes in osmotic processes: a review. Clays and Clay Minerals, 34 (2), 214-223.

FRITZ, S.J. and EADY, C.D. (1985): Hyperfiltration-induced precipitation of calcite. Geochimica et Cosmochimica Acta, 49, 761-768.

FRITZ, S.J. and MARINE, I.W. (1983): Experimental support for a predictive osmotic model of clay membranes. Geochimica et Cosmochimica Acta, 47, 1515-1522.

GARRELS, R.M. and CHRIST, C.L (1965): Solutions, minerals and equilibria. Freeman, Cooper & Company, USA.

GAUTSCHI, A. (1989): Geology and hydrogeology of northern Switzerland. Proc. 6th Int. Symp. on Water-Rock Interaction, Malvern, Publ. A.A. Balkema, Rotterdam, 255-258.

GIBSON, R.E. (1958): The progress of consolidation in a clay layer increasing in thickness with time. Geotechnique, 8, 171-182.

GOUY, G. (1910): Sur la constitution de la charge électrique à la surface d'un électrolyte. J. Physique, 9, 457-468.

GOUY, G. (1917): Sur la fonction électrocapillaire. Ann. Phys. (Paris), Séries 9, 7, 129-184.

GRAF, D.L., MEENTS, W.F., FRIEDMAN, T. and SHIMP, N.F. (1966): The origin of saline formation waters; Calcium chloride waters. Illinois State Geol. Survey Circ., 397.

GRAF, D.L. (1982): Chemical osmosis, reverse chemical osmosis, and the origins of subsurface brines. Geochimica et Cosmochimica Acta, 46, 1431-1448.

GRAY, G.R., DARLEY, H.C.H., and ROGERS, W.F. (1980): Composition and properties of oil well drilling fluids. Gulf Pub. Co.

GREENBERG, J.A. (1971): Diffusional flow of salt and water in soils. Ph.D. Thesis, University of California, Berkely.

GREENBERG, J.A., MITCHELL, J.K. and WITHERSPOON, P.A. (1973): Coupled salt and water flows in a groundwater basin. J. Geophys. Res., 78, 6341-6353.

GROB, H. (1976): Swelling and heave in Swiss tunnels. Bull. Int. Assoc. Engng. Geol., 13, 55-60.

HANNA, T.H. (1973): Foundation instrumentation. Trans Tech Publications, Aedermannsdorf, Switzerland.

HARDY, R.M. (1965): Identification and performance of swelling soil types. Canadian Geotech. J., 11(2).

HAYDON, P.R. and GRAF, D.L. (1986): Studies of smectite membrane behavior: Temperature dependence, 20-180°C. Geochimica et Cosmochimica Acta, 50, 115-121.

HITCHON, B., BILLINGS, G.K. and KLOVAN, J.E. (1971): Geochemistry and origin of formation waters in the western Canada sedimentary basin - III: Factors controlling chemical composition. Geochimica et Cosmochimica Acta, 35, 567-598.

HOBBS, P.R.N., YEOW, H., HORSEMAN, S.T. and JACKSON, P.D. (1982): Swelling properties of the mudrocks at Harwell, Rept. Institute of Geological Sciences (NERC), ENPU 82-11.

HUANG, S.L., AUGHENBAUGH, N.B. and ROCKAWAY, J.D. (1986): Swelling pressure studies of shales. Int. J. Rock Mech Min. Sci., & Geomech. Abstr., 23(5), 371-377.

HUBBERT, M.K (1940): The theory of groundwater motion. J. Geol., 48, 785-944.

JONES, P.H. (1968): Geochemical hydrodynamics - a possible key to the hydrology of certain aquifer systems in the northern part of the gulf of Mexico basin. XXIII International Geological Congress, 17, 113-125.

KATCHALSKY, A. and CURRAN, P.R. (1967): Nonequilibrium thermodynamics in biophysics. Harvard University Press, Cambridge, Mass.

KELLY, J. Jr. (1969): Drilling problem shales. Oil and Gas Journal, 9, 21-35.

KIMMEIER, F., PERROCHET, P., ANDREWS, R. and KIRALY, L. (1985): Simulation par modèle mathématique des écoulements souterrains entre les Alpes et la Forêt Noire. Rapport Technique NTB 84-50, NAGRA, Baden Switzerland.

KOJIMA, K., IKEDA, K. and KAWAI, K. (1977): Mud compaction and the mathematical model for "compaction current" in sedimentary basin. Japan Assoc. Pet. Tech. J., 42, 100-106.

KOPPULA, S.D. (1983): Porewater pressure due to overburden removal. J. Geotech. Eng., A.S.C.E., 109, 1099-1110.

KOPPULA, S.D. and MORGENSTERN, N.R. (1984): Deficient pore pressures in an eroding soil mass. Canadian Geotechnical J., 21, 277-288.

KOVARI, K., MADSEN, F.T. and AMSTAD, C. (1981): Tunnelling with yielding support in swelling rocks. Proc. Int. Symp. Weak Rock, I.S.R.M. Tokyo, 2, 1-19-1026.

LAMA R.D. and VUTUKURI, V.S. (1978): Handbook on mechanical properties of rock: Testing techniques and results, (II), Trans Tech Publications, Aedermannsdorf, Switzerland.

LAMBE, T.W. (1960): A mechanistic picture of shear strength in clay. Proc. Res. Conf. on Shear Strength of Cohesive Soils, A.S.C.E., 437.

LANGMUIR, I. (1938): The role of attractive and repulsive forces in the formation of tactoids, thixiotropic gels, protein crystals, and coacervates. J. Chem. Phys., 6, 873-896.

LETEY, J. and KEMPER, W.D. (1969): Movement of water and salt through a clay-water system: experimental verification of Onsager reciprocal relation. Soil Science Socity of America, Proceedings, 33, 25-29.

LEWIS, C.R. and ROSE, S.C. (1970): A theory relating high temperatures and overpressures. J. Pet. Technol., 22, 11-16.

LOW, P.F. (1961): Physical chemistry of clay-water interaction. Advances in Agronomy, 13, 269-327.

LOW, P.F. (1987): The clay-water interface. Proc. Int. Clay Conf., Denver, The Clay Minerals Society, Bloomington, Indiana, 247-256.

MADSEN, F.T. (1976): Quelldrucksmessung an Tongesteinen und Berechnung des Quelldruckes nach der DVLO-Theorie. Mitteilungen des Institutes fuer Grundbau und Bodenmechanik Nr. 108, Eidgenoess. Techn. Hochschule Zuerich.

MADSEN, F.T. and MULLER-VONMOOS, M. (1985): Swelling pressure calculated from minerological properties of a Jurassic Opalinum shale, Switzerland. Clays and Clay Minerals, 33, 501-509.

MAGARA, K. (1974): Aquathermal fluid migration. Bull. Am. Assoc Pet. Geol., 58, 283-290.

MAGARA, K. (1975): Importance of aquathermal pressuring effect in Gulf Coast. Bull. Am. Assoc. Pet. Geol., 59, 2037-2045.

MARINE, W.I. and FRITZ, S.J. (1981): Osmotic model to explain anomalous hydraulic heads. Water Resources Research, 17(1), 73-82.

MARTIN, R.T. (1962): Adsorbed water on a clay: a review. Clays and Clay Minerals, 9 (Proceedings of the 9th National Conf. Clays and Clay Minerals, 1960), Pergamon, New York, 28-270.

MITCHELL, J.K. (1976): Fundamentals of Soil Behavior. John Wiley and Sons, New York.

MITCHELL, J. K. and YOUNGER, J. S. (1967): Abnormalities in hydraulic flow through fine-grained soils. American Society Testing Materials, Special Publication 417, 106-141.

MITCHELL, J.K., GREENBERG, J.A. and WITHERSPOON, P.A. (1973): Chemico-osmotic effects in fine-grained soils. J. Soil Mech. and Found. Div., A.S.C.E., 99, 307-322.

NAGRA (1988): Sedimentstudie (Zwischenbericht), Technical Report NTB 88-25, Nationale Genossenschaft für die Lagerung radioactiver Abfälle (NAGRA), Baden, Switzerland.

NEUZIL, C.E. (1986): Groundwater flow in low-permeability environments. Water Resources Research, 22, 1163-1195.

NEUZIL, C.E. and POLLOCK, D.W. (1983): Erosional unloading and fluid pressures in hydraulically "tight" rocks. J. Geol., 91(2), 179-193.

NUESCH, R. (1988): Felsmechanische Resultate aus Untersuchungen an Opalinuston. Internal Report, Institute for Soil and Rock Mechanics, E.T.H., Zürich, prepared for Nationale Genossenschaft für die Lagerung radioactiver Abfälle (NAGRA), Baden, Switzerland.

NUR, A. and BYERLEE, J.D. (1971): An exact effective stress law for elastic deformation of rock with fluids. J. Geophys. Res, 76, 6414-6419.

OLSEN, H. W. (1960): Hydraulic flow through saturated clays. 9th National Conference on Clays and Clay minerals, Lafayette, pp 131-161.

OLSEN, H. W (1965): Deviations from Darcy's law in saturated clays. Proceedings American Soil Science Society, 29 (2), 135-140.

OLSEN, H. W. (1966): Darcy's law in saturated kaolinite. Water Resources Research, 2(2), 287-295.

OLSEN, H. W. (1969): Simultaneous fluxes of liquid and change through saturated kaolinite. Proceedings Soil Science Society America, 33(3), 338-344.

OLSEN, H.W. (1972): Liquid movement through kaolinite under hydraulic, electric and osmotic gradients. Bulletin American Association Petroleum Geology, 56(10), 2022-2028.

OLSEN, H. W. (1985): Osmosis: a cause of apparent deviations from Darcy's Law. Canadian Geotechnical Journal, 22, 238-241.

PALCIAUSKAS, V.V. and DOMENICO, P.A. (1989): Fluid pressures in deforming porous rocks. Water Resources Research, 25, 203-213.

POWERS, M.C. (1967): Fluid-release mechanisms in compacted marine mudrocks and their importance in oil exploration. Bull. Am. Assoc. Geol., 51, 1240-1254.

RAVINA, I. and ZASLAVSKY, D. (1972): The water pressure in the electrical double layer. Israel J. Chemistry, 10, 707-714.

RIEKE, H.H. and CHILINGARIAN, G.V. (1974): Compaction of argillaceous sediments. Elsevier, New York, 424.

ROBINS, N.S., MARTIN, B.A. and BRIGHTMAN, M.A. (1981): Borehole drilling and completion: Details for the Harwell research site. Rept. Institute of Geological Sciences (NERC), ENPU 81-9.

RUBEY, W.W. and HUBBERT, M.K. (1959): Role of fluid pressure in mechanics of overthrust faulting, II. Geol. Soc. Am. Bull., 70, 167-206.

RUSSELL, W.L. (1972): Pressure-depth relations in Appalachian region. Bull. Am. Assoc. Pet. Geol., 56, 528-536.

SCHAER, J.P. (1979): Mouvements verticaux, érosion dans les Alpes, aujourd'hui et au cours du Miocène. Eclogae geol. Helv., 72(1), 263-270.

SCHAER, J.P. and JEANRICHARD, F. (1974): Mouvements verticaux anciens et actuels dans les Alpes suisses. Eclogae geol. Helv., 67(1), 101-119.

SCHAER, J.P., REIMER, G.M. and WAGNER, G.A. (1975): Actual and ancient uplift rate in the Gotthard region, Swiss Alps: A comparison between precise levelling and fission-track aplite age. Tectonophysics, 29, 293-300.

SCHOFIELD, A. and WROTH, P.W.(1968): Critical state soil mechanics. McGraw-Hill, London.

SERATA, S. and GLOYNA, E.F. (1960): Principles of structural stability of underground salt caverns. J. Geophys. Res., 65(9), 2979-2987.

SHI, Y. and WANG, C. (1986): Pore pressure generation in sedimentary basins: overloading versus aquathermal. J. Geophys. Res., 91, 2153-2162.

SMITH, J.E. (1971): The dynamics of shale compaction and evolution of pore fluid pressure. Int. Assoc. Math. Geol. J., 3, 239-263.

STAVERMAN, A.J. (1952): Non-equilibrium thermodynamics of membrane processes. Trans. Faraday Society, 48, 176-185.

STERN, O. (1924): Zur Theorie der Elektrolytischen Doppelschicht. Zeitschrift Electrochem, 30, 508-516.

TAYLOR, D.W. (1948): Fundamentals of soil mechanics. Wiley, New York.

TOTH, J. and MILLAR, R.F. (1983): Possible effects of erosional changes of the topographic relief on pore pressures at depth. Water Resources Research, 19(6), 1585-1597.

THURY, M. and DIEBOLD, P. (1987): Ueberblick ueber das geologische Untersuchungsprogram der NAGRA in der Nordschweiz. Eglogae geol. Helv., 80 (2) 271-286.

VAN OLPHEN, H. (1977): An introduction to clay colloid chemistry. Wiley-Interscience, New York.

VERRUIJT, A. (1984): The theory of consolidation. *In :* Fundamentals of transport phenomena in porous media, Edited by J. Bear and M.Y. Corapcioglu, Proc. NATO Advanced Study Institute on Mechanics of Fluids in Porous Media, Delaware, USA.

VERWEY, E.J.W. and OVERBEEK, J. Th. G. (1948): Theory of the stability of lyophobic colloids. Elsevier, Amsterdam.

VON ENGELHARDT, W. (1960): Der Porenraum der Sedimente. Springer, Berlin.

WICHTER, L. (1979): On the geotechnical properties of a Jurassic clay shale. Proc. 4th Cong. Int. Soc., Rock Mech., Montreux, Switzerland.

WROTH, C.P. (1972): Some aspects of the elastic behaviour of overconsolidated clay, *In:* Stress strain behaviour of soils, Roscoe Memorial Symposium, Cambridge, Edited by R.G.H. Parry, Publ. G.T. Fowlis, Henley-on-Thames, 347-361.

YONG, R.N and WARKENTIN, B.P. (1975): Soil properties and behaviour. Developments in Geotechnical Engineering, 5, Elsevier, Amsterdam.

UNCITED REFERENCES

AITCHISON, G.D., RUSSAM, D. and RICHARDS, B.G. (1965): Engineering concepts of moisture equilibrium and moisture changes in soils. Moisture Equilibrium and Moisture Changes in Soils Beneath Covered Areas, Butterworths, Sidney, Australia.

BEAR, J. (1972): Hydraulics of groundwater. Series in water resources and environmental engineering, McGraw-Hill, New York.

BERRY, F.A.F. (1960): Geologic field evidence suggesting membrane properties of shales. Bulletin American Association Petroleum Geology, 44(6), 953-954.

BREDERHOEFT, J.D., NEUZIL, C. E., and MILLEY, P. C. D. (1983): Regional flow in the Dakota Aquifer: A study of the role of confining layers. U. S. Geological Survey, Water Supply Paper 2237, 1-45.

CARNAHAN, C.L. (1987): Effects of coupled thermal, hydrogeological and chemical processes on nuclide transport. Proc. of Geoval 87, Stockholm, Sweden.

DEMIR, I. (1988): Studies of smectite membrane behavior: Electrokinetic, osmotic, and isotopic fractionation processes at elevated pressures. Geochimica et Cosmochima Acta, 52, 727-737.

FERTL, W.H. (1976): Abnormal Formation Pressures - Implications to Exploration, Drilling, and Production of Oil and Gas Resources. Developments in Petroleum Science, 2, Elsevier, Amsterdam.

HANSHAW, B.B. (1964): Cation-exchange constants for clays from electro-chemical measurements. *In:* Clays and clay minerals: Bradley, W. F. (editor), Pergamon Press, New York, p. 397-421.

HANSHAW, B.B. and ZEN, E. (1965): Osmotic equilibrium and overthrust faulting. Bulletin of the Geological Society of America, 76, 1379-1387.

HANSHAW, B.B. (1972): Natural-membrane phenomena and subsurface waste emplacement. *In:* Underground Waste Management and Environmental Implications, ed. T.D. Cook, Memoirs American Association Petroleum Geology, 18, 308-317.

HITCHON, B. and FRIEDMAN, I. (1969): Geochemistry and origin of formation waters in the western Canada sedimentary basin - 1. Stable isotopes of hydrogen and oxygen. Geochimica et Cosmochimica Acta, 33, 1321-1349.

HUBBERT, M.K. and RUBEY, W.W. (1959): Role of fluid pressure in mechanics of overthrust faulting. I. Mechanics of fluid-filled porous solids and its application to overthrust faulting. Bull. Geol. Soc. America, 70, 115-166.

JORGENSEN, D.G. (1980): Relationships between basic soils engineering equations and basic ground water flow equations. U.S. Geol. Surv., Water Supply Paper 2064.

MAGARA, K. (1978): Compaction and Fluid Migration - Practical Petroleum Geology. Developments in Petroleum Science, 9, Elsevier, Amsterdam.

MARINE, W.I. (1974): Geohydrology of the buried Triassic basin at Savannah river plant, South Carolina. Bull. Am. Assoc. Pet. Geol., 58 (9), 1825-1837.

OLSEN, S.R. and KEMPER, W.D. (1968): Movement of nutrients to plant roots. Advances in Agronomy, 20, 91-151.

PHILLIPS, F.M. (1983): Comment on "Chemical osmosis, reverse chemical osmosis, and the origin of subsurface brines" by Donald L. Graf. Geochimica et Cosmochimica Acta, 47, 1331.

NOTATION

a	Borehole radius
a	Activity
$\mathbf{a}_A$, $\mathbf{a}_B$	Activity of solutions in beds A and B, respectively
A	Pore pressure coefficent
A	Attractive force per unit area
B	Pore pressure coefficient
C	Clay fraction

C	Cation exchange capacity
C_A, C_B	Solute concentration in beds A and B, repectively
C_p	Solute concentration in the macropores
C_S	Solute concentration
C_0	Solute concentration of external solution
C_0^+, C_0^-	Concentration of ions in external solution
C_m^+, C_m^-	Concentration of ions at the midplane
C_{sm}	Maximum value of solute concentration
C_v	Coefficient of consolidation
d	Half distance between clay platelets
D	Dielectric constant of water
D	Diffusion coefficient
D'	Combined coefficient of coupled flow theory
e	Voids ratio
e	Elementary charge
e_1, e_2	Voids ratio constants
e_a	Voids ratio at reference stress on VCL
e_b	Voids ratio at reference stress on RRL
E	Young's modulus (of elasticity)
E_k	Constrained modulus (of elasticity)
E_u	Undrained modulus (of elasticity)
g	Gravitational constant
G_S	Vertical total stress gradient
G_T	Geothermal gradient
h	Total hydraulic head
$\mathbf{h}_0$, $\mathbf{h}_1$, etc.	Total hydraulic head at prescribed points
h_p	Pressure head
h_{pmeas}	Pressure head measured during testing
h_{p0}, h_{p1}, etc.	Pressure head at prescribed points
h_{pm}	Pressure head at the midplane
h_{pp}	Pressure head in a macropore
h_s	Solute head
h_{s0},h_{s1}, etc.	Solute head at prescribed points
h_{sm}	Solute head at the midplane
h_{sp}	Solute head in a macropore
H	Biot's constant

H	Half thickness of clay stratum
i, j, k	Range variables
J_i	Flows of coupled flow theory
J_D	Flow rate of solute relative to water
J_W	Flow rate of water
k	Boltzmann's constant
K	Bulk modulus (of elasticity)
$\mathbf{K}$	Hydraulic conductivity
$\underset{\approx}{\mathbf{K}}$	Hydraulic conductivity tensor
$\mathbf{K}_{ch}$	Coupling coefficient
$\mathbf{K}_{hc}$	Coupling coefficient
$\boldsymbol{K}_{ch}$	Combined coefficient of coupled flow theory
K_s	Bulk modulus of elasticity of constituent minerals (solids)
$\mathbf{K}_v$	Vertical hydraulic conductivity
$\mathbf{K}_{vf}$	Vertical hydraulic conductivity of fractured rock mass
L_{ij}	Phenomenological coefficients of coupled flow theory
m	Water content
m_v	One-dimensional compressibility (reciprocal of E_k)
M	Variable in consolidation theory
n	Porosity
n	Ion concentration
n_0	Ion concentration at a particular point
N_A	Avogadro's number
N_s	Number of moles of solute per unit volume of clay
N_w	Number of moles of water per unit volume of clay
P	Osmotic pressure
P_{bore}	Pressure in the borehole
$\underset{\sim}{q}$	Specific discharge (vector)
$\underset{\sim}{q_r}$	Specific discharge relative to moving solid (vector)
$\underset{\sim}{Q}$	Biot's constant
r	Radial distance
R	Biot's constant
$\mathbf{R}$	Gas constant
R	Osmotic repulsive force per unit area, referenced to pure water
R_{diff}	Osmotic repulsive force per unit area, referenced to specified water

RRL	Rebound/Reconsolidation Line (abbrev.)
S	Specific surface area (of clay minerals)
S_u	Undrained shear strength
t	Time
t_{95}	Time for 95% excess pore pressure dissipation
T	Temperature
T_0	Reference temperature
T_v	Non-dimensional time
u	Pore pressure
u	Substituted variable
u_{ex}	Excess pore pressure relative to hydrostatic
u_p	Water pressure in a macropore
u_0	Water pressure in a piezometer containing pure water
v	Velocity of groundwater
$\underset{\sim}{v}_s$	Velocity of solids (vector)
$\underset{\sim}{v}_w$	Velocity of water (vector)
V_w	Partial molar volume of water
VCL	Virgin Consolidation Line (abbrev.)
x	Distance variable
X_j	Potential gradients (coupled flow theory)
y	Substituted variable
z	Depth variable *and* Elevation head
z	Substituted variable
α_b	Bulk thermal expansion coefficient (cubical) of rock
α_l	Linear thermal expansion coefficient of rock
α_m	Thermal expansion coefficient (Biot's theory)
α_s	Thermal expansion coefficient (cubical) of solids
α_w	Thermal expansion coefficient of water
β	Bulk compressibility
β_p	Pore compressibility
β_s	Compressibility of mineral constituents (solids)
β_s'	Compressibility term
β_w	Compressibility of water
ε	Volumetric strain
ε_{11}, ε_{22}, ε_{33}	Principal strain components
ζ	Biot's constant

γ_s	Density of mineral constituents (solids)
γ_w	Density of water
ϕ	Total potential of groundwater
ϕ_p	Pressure potential
ϕ_s	Solute potential
ϕ_z	Elevation potential
ϕ^*	Hubbert's potential for a compressible fluid
κ	- Slope of RRL on [e, ln σ'] plot
λ	- Slope of VCL on [e, ln σ'] plot
ν	Poisson's ratio
ν	Valence
ρ	Net space charge density
ξ	Electrical field intensity
Π	Osmotic pressure (theoretical)
σ	Stress
σ	Surface charge on clay platelet
σ'	Effective stress
σ_i'	Interparticle stress
σ_m	Mean normal total stress
σ_m'	Mean normal effective stress
σ_r	Radial stress (total)
σ_t	Tangential stress (total)
σ_v	Vertical stress (total)
σ_v'	Vertical stress (effective)
σ_z	Axial stress (total)
σ_{11}, σ_{22}, σ_{33}	Principal stress components (total)
χ	Substituted variable
ψ	Electrical potential
ψ_m	Electrical potential at the midplane
ψ_s	Electrical potential at the clay particle surface

Mathematical Notation

$$\nabla\cdot(\ ) = \frac{\partial(\ )}{\partial x} + \frac{\partial(\ )}{\partial y} + \frac{\partial(\ )}{\partial z}$$ Divergence operator

$$\text{grad}(\ ) \equiv \frac{\partial(\ )}{\partial x}\underset{\sim}{\mathbf{i}} + \frac{\partial(\ )}{\partial y}\underset{\sim}{\mathbf{j}} + \frac{\partial(\ )}{\partial z}\underset{\sim}{\mathbf{k}}$$ Gradient

$$\frac{D_w(\ )}{Dt} = \frac{\partial(\ )}{\partial t} + \underset{\sim}{\mathbf{v}}_w \nabla\cdot(\ )$$ Material derivative

APPENDIX A

Pore Water Pressures in a Deforming Porous Medium with Temperature Variations

The equation of water mass conservation in a saturated porous medium is

$$\nabla \cdot \gamma_w \underset{\sim}{q} + \frac{\partial(\gamma_w n)}{\partial t} = 0 \qquad \underset{\sim}{q} = n \underset{\sim}{v}_w \tag{A1}$$

where $\underset{\sim}{v}_w$ and $\underset{\sim}{q}$ are the water's velocity and specific discharge, respectively, and γ_w is the density of water (BEAR, 1972).

In a porous deforming medium it is the specific discharge relative to the moving solid q_r that is expressed by Darcy's law

$$\underset{\sim}{q}_r = \underset{\sim}{q} - n \underset{\sim}{v}_s = - \underset{\approx}{\mathbf{K}} \cdot \nabla \phi^* \tag{A2}$$

where v_s is the velocity of the solid, $\underset{\approx}{\mathbf{K}}$ is the hydraulic conductivity tensor and $\phi*$ is Hubbert's potential for a compressible fluid

$$\phi^* = z + \int_{u_o}^{u} \frac{du}{g\,\gamma_w(u)} \tag{A3}$$

The equation of solid mass conservation is

$$\nabla \cdot [\, \gamma_s (1-n) \underset{\sim}{v}_s \,] + \frac{\partial[\, \gamma_s (1-n) \underset{\sim}{v}_s \,]}{\partial t} = 0 \tag{A4}$$

where γ_s is the density of the solid particles.

The material derivatives with repect to the water and the solids are defined as

$$\frac{D_w(\ )}{Dt} = \frac{\partial(\ )}{\partial t} + \underset{\sim}{v}_w \cdot \nabla(\ ) \tag{A5}$$

$$\frac{D_s(\ )}{Dt} = \frac{\partial(\ )}{\partial t} + \underset{\sim}{v}_s \cdot \nabla(\ ) \tag{A6}$$

Inserting (A2) into (A1) and using (A5) we obtain

$$\gamma_w \nabla \cdot \underset{\sim}{q}_r + n \frac{D_w \gamma_w}{Dt} + \gamma_w \frac{D_s n}{Dt} + \gamma_w n \nabla \cdot \underset{\sim}{v}_s = 0 \tag{A7}$$

Using (A4) and (A6) we obtain

$$\frac{D_s n}{Dt} = \frac{(1-n)}{\gamma_s} \frac{D_s \gamma_s}{Dt} + (1-n) \nabla \cdot \underset{\sim}{v}_s \tag{A8}$$

Substituting (A8) in (A7) gives

$$\nabla \cdot \underset{\sim}{q}_r + \nabla \cdot \underset{\sim}{v}_s + \frac{n}{\gamma_w} \frac{D_w \gamma_w}{Dt} + \frac{(1-n)}{\gamma_s} \frac{D_s \gamma_s}{Dt} = 0 \tag{A9}$$

Letting vector **u** denote the solid's displacement and ε the volumetric strain, then

$$\varepsilon = -\nabla \cdot \underset{\sim}{u} = -\left[\frac{\partial \underset{\sim}{u}_x}{\partial x} + \frac{\partial \underset{\sim}{u}_y}{\partial y} + \frac{\partial \underset{\sim}{u}_z}{\partial z} \right] \tag{A10}$$

Thus the velocity of the solid phase is

$$v_s = \frac{\partial \underset{\sim}{u}}{\partial t} \tag{A11}$$

From which follows

$$\nabla \cdot \underset{\sim}{v_s} = - \frac{D_s \varepsilon}{Dt} \qquad \text{(A12)}$$

Substituting (12) in (9) we obtain one particular form for the three-dimensional equation of mass conservation

$$\nabla \cdot \underset{\sim}{q_r} - \frac{D_s \varepsilon}{Dt} + \frac{n}{\gamma_w} \frac{D_w \gamma_w}{Dt} + \frac{(1-n)}{\gamma_s} \frac{D_s \gamma_s}{Dt} = 0 \qquad \text{(A13)}$$

Assuming $|\partial\varepsilon/\partial t| >> |\underset{\sim}{v_s} \nabla\cdot\varepsilon|$, $|\partial\gamma_w/\partial t| >> |\underset{\sim}{v_w} \nabla\cdot\gamma_w|$ and $|\partial\gamma s/\partial t| >> |\underset{\sim}{v_s} \nabla\cdot\gamma_s|$ we can re-write (A13) in terms of the partial derivatives.

$$\nabla \cdot \underset{\sim}{q_r} - \frac{\partial \varepsilon}{\partial t} + \frac{n}{\gamma_w} \frac{\partial \gamma_w}{\partial t} + \frac{(1-n)}{\gamma_s} \frac{\partial \gamma_s}{\partial t} = 0 \qquad \text{(A14)}$$

Our purpose is to express (A14) in terms of the partial derivatives with respect to time of pore pressure, total stress and temperature.

Changes in water density may be described by

$$\frac{d\gamma_w}{\gamma_w} = \beta_w \, du - \alpha_w \, dT \qquad \text{(A15)}$$

where u is pore pressure, T is temperature, β_w is the isothermal compressibility of water and α_w is the isobarometric thermal expansion coefficient.

Difficulties arise in developing an equivalent expression for density changes in the solids, due primarily to the inhomogenous deformation of individual grains when the porous medium is subject to stress.

Identifying the appropriate stress term to be the mean normal total stress σ_m where

$$\sigma_m = \frac{1}{3}(\sigma_{11} + \sigma_{22} + \sigma_{33}) \tag{A16}$$

and taking a linear approximation, PALCIAUSKAS and DOMENICO (1989) obtain

$$(1-n)\frac{d\gamma_s}{\gamma_s} = \beta_s \, d\sigma_m - n\,\beta_s' \, du - (1-n)\,\alpha_s \, dT \tag{A17}$$

where β_S and β_S' are two independent grain compressibilities defined by

$$\beta_s = \frac{1}{K_s} = \frac{(1-\nu)}{\gamma_s}\left(\frac{\partial \gamma_s}{\partial \sigma_m}\right)_{u,T} \tag{A18}$$

and

$$\beta_s' = -\frac{(1-n)}{n\,\gamma_s}\left(\frac{\partial \gamma_s}{\partial u}\right)_{\sigma_m,T} \tag{A19}$$

and α_s is the coefficient of cubical thermal expansion of the grains

$$\alpha_s = -\frac{1}{\gamma_s}\left(\frac{\partial \gamma_s}{\partial T}\right)_{u,\sigma_m} \tag{A20}$$

Using (A15) and (A17), we obtain for (14)

$$n(\beta_w - \beta_s')\frac{\partial u}{\partial t} = -\nabla \cdot \underset{\sim}{q_r} + \frac{\partial \varepsilon}{\partial t} - \beta_s \frac{\partial \sigma_m}{\partial t} + (n\,\alpha_w + (1-n)\,\alpha_s)\frac{\partial T}{\partial t} \tag{A21}$$

We now require to evaluate the $\partial\varepsilon/\partial t$ term by introducing a constitutive equation describing the deformation of the particular material. We recognize that the volumetric deformation of clays and shales is significantly nonlinear with respect to stress and we

might anticipate similar nonlinearity in the response to temperature. Additional complications are non-reversible or "plastic" deformation, time-dependent deformation or "creep", and possible sensitivity of the volumetric strains to the deviatoric (shear) conponents of stress. In theory we could introduce a variety of different constitutive equations in (A21). For the time being we restrict ourselves to isotropic, linear elastic elasticity. In doing so we must be aware that the theory only holds for small strains and that the material constants must be chosen carefully to be representative not only of the stress and temperature regime of the problem under consideration but also of the time period.

Theoretical background on the development of a constitutive equation describing the stress-strain behaviour of an porous elastic medium is provided by NUR and BYERLEE (1971), amongst others.

Ignoring deviatoric components, the equation can be written in the form

$$\varepsilon = \beta \sigma_m - (\beta - \beta_s) u - \alpha_b (T - T_0) \qquad \text{(A22)}$$

where β is the bulk compressibility of the porous rock

$$\beta = \frac{1}{K} = -\frac{1}{V} \left(\frac{\partial V}{\partial \sigma_m} \right)_{u,T} \qquad \text{(A23)}$$

It is assumed that the coefficient of cubical thermal expansion of the rock α_b can be expressed by

$$\alpha_b = (1 - n) \alpha_s + n \alpha_p \qquad \text{(A24)}$$

where α_p is the thermal expansion coefficient of the pores.

Using (A22) in incremental form and applying it in (A21) gives

$$(\beta - \beta_s - n\beta_s' + n\beta_w)\frac{\partial u}{\partial t} = -\nabla\cdot\underset{\sim}{q_r} + (\beta - \beta_s)\frac{\partial \sigma_m}{\partial t} + (n\alpha_w + (1-n)\alpha_s - \alpha_b)\frac{\partial T}{\partial t} \quad \text{(A25)}$$

Identifying $(\beta - \beta_s - n\,\beta_s')$ as the pore compressibility β_p and using (A2), (A24) and (A25) we obtain

$$(\beta_p + n\beta_w)\frac{\partial u}{\partial t} = \nabla\cdot(\underset{\approx}{\mathbf{K}}\,\nabla\cdot\phi^*) + (\beta - \beta_s)\frac{\partial \sigma_m}{\partial t} + n(\alpha_w - \alpha_p)\frac{\partial T}{\partial t} \quad \text{(A26)}$$

or, in terms of Biot's constants

$$\frac{1}{R}\frac{\partial u}{\partial t} = \nabla\cdot(\underset{\approx}{\mathbf{K}}\,\nabla\cdot\phi^*) + \frac{1}{H}\frac{\partial \sigma_m}{\partial t} + \alpha_m\frac{\partial T}{\partial t} \quad \text{(A27)}$$

Eliminating the stress term in (A21) using (A22) we find

$$[(\beta_p + n\beta_w) - (\frac{\beta_s^2}{\beta} + \beta + 2\beta_s)]\frac{\partial u}{\partial t} = \nabla\cdot(\underset{\approx}{\mathbf{K}}\,\nabla\cdot\phi^*) + (1 - \frac{\beta_s}{\beta})\frac{\partial \varepsilon}{\partial t} + (\alpha_b(1 - \frac{\beta_s}{\beta}) + n(\alpha_w - \alpha_p))\frac{\partial T}{\partial t} \quad \text{(A28)}$$

or, in terms of Biot's constants

$$(\frac{1}{R} - \frac{K}{H^2})\frac{\partial u}{\partial t} = \nabla\cdot(\underset{\approx}{K}\,\nabla\cdot\phi^*) + \frac{K}{H}\frac{\partial \varepsilon}{\partial t} + (\alpha_b\frac{K}{H} + \alpha_m)\frac{\partial T}{\partial t} \quad \text{(A29)}$$

which can also be expressed as

$$\frac{1}{Q}\frac{\partial u}{\partial t} = \nabla\cdot(\underset{\approx}{\mathbf{K}}\,\nabla\cdot\phi^*) + \zeta\frac{\partial \varepsilon}{\partial t} + (\zeta\alpha_b + \alpha_m)\frac{\partial T}{\partial t} \quad \text{(A30)}$$

APPENDIX B

One-Dimensional Transient Flow in Clays under Combined Hydraulic and Chemical Gradients

An isotropic, homogenenous, horizontal, saturated clay layer is considered under isothermal conditions. It is assumed that no ion exchange or adsorption reactions occur during diffusion.

From the postulates of irreversible thermodynamics equations for the flow rate of water J_w and for the flow rate of solute relative to the water J_D can be derived (KATCHALSKY and CURRAN, 1967; LETEY and KEMPER, 1969).

$$J_w = L_{11} V_w \ \mathrm{grad}(-u) + L_{12}(\mathbf{R}T/C_S) \ \mathrm{grad}(-C_S) \qquad \text{(B1)}$$

$$J_D = L_{21} V_w \ \mathrm{grad}(-u) + L_{22}(\mathbf{R}T/C_S) \ \mathrm{grad}(-C_S) \qquad \text{(B2)}$$

where L_{ij} represents phenomenological coefficients, V_w is the partial molar volume of water, u is the hydrostatic pressure, **R** is the gas constant, T is the absolute temperature and C_S is the molar solute concentration. According to Onsager's reciprocity theorem,

$$L_{12} = L_{21} \qquad \text{(B3)}$$

The flow of solute relative to the fixed soil layer J_S is of more significance than that relative to the water J_D. They are related by

$$J_S = J_D + (C_S/C_w) \cdot J_w \qquad \text{(B4)}$$

where $C_w = 1/V_w$ = molar concentration of water.

By the principle of the conservation of mass, the rate of increase of mass density at any point must equal the rate at which matter flows to the point.

For water in the pores

$$\nabla \cdot J_w = - \frac{\partial N_w}{\partial t} \tag{B5}$$

and for solute

$$\nabla \cdot J_S = - \frac{\partial N_S}{\partial t} \tag{B6}$$

where N_w and N_S are the number of moles of water and solute, respectively, per unit volume of clay. Combination of the flow equations with the continuity equations leads, after replacement of J_D, to

$$- \frac{\partial N_w}{\partial t} = \nabla \cdot \left[L_{11} V_w \text{ grad}(-u) + L_{12} (\mathbf{R}T/C_s) \text{ grad}(-C_s) \right] \tag{B7}$$

$$\begin{aligned} - \frac{\partial N_S}{\partial t} = \nabla \cdot [\{ L_{21} V_w + (C_s/C_w) L_{11} V_w \} \text{ grad}(-u) \\ + \{ L_{22} (\mathbf{R}T/C_s) + L_{12} (\mathbf{R}T/C_w) \} \text{ grad}(-C_s)] \end{aligned} \tag{B8}$$

The phenomenological coefficients are given by

$$L_{11} = \frac{\mathbf{K}}{V_w^2 \gamma_w g} \tag{B9}$$

$$L_{12} = \frac{C_s}{\mathbf{R}T V_w} \mathbf{K}_{hc} \tag{B10}$$

$$L_{22} = \frac{C_s}{\mathbf{R}T} \mathbf{D} \tag{B11}$$

$$L_{21} = \frac{(C_s/C_{sm})}{V_w \gamma_w g} \mathbf{K}_{ch} \tag{B12}$$

where γ_w is the density of water, **D** is the diffusion coefficient of the solute, $\mathbf{K}_{hc}$ and $\mathbf{K}_{ch}$ are the coupling coefficients, C_{sm} is the maximum value of C_s, and **K** is the hydraulic conductivity.

Assuming that the compression behaviour of the clay is linear

$$\Delta e = \frac{(1+e)}{E_k} \Delta u \tag{B13}$$

where e is the voids ratio and E_k is the constrained modulus. Taking z to be the coordinate direction, the one-dimensional form of the diffusion equations become

$$\frac{1}{C_v} \frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial z^2} + \frac{\mathbf{K}_{hc} \gamma_w g}{\mathbf{K}} \cdot \frac{\partial^2 C_s}{\partial z^2} \tag{B14}$$

and

$$n \frac{\partial C_s}{\partial t} = \frac{\boldsymbol{K}_{ch}}{\gamma_w g} \cdot \frac{\partial}{\partial z}((C_s/C_{sm})\frac{\partial u}{\partial z}) + \mathbf{D}' \frac{\partial^2 C_s}{\partial z^2} - \frac{C_s}{E_k} \frac{\partial u}{\partial t} \tag{B15}$$

where porosity n = e/(1+e),

$$\boldsymbol{K}_{ch} = \mathbf{K}_{ch} + C_{SM}\, \mathbf{K} \tag{B16}$$

$$\mathbf{D}' = \mathbf{D} + C_S\, \mathbf{K}_{hc} \tag{B17}$$

and $C_v = (E_k\, \mathbf{K})/(\gamma_w\, g)$, the coefficient of consolidation.

APPENDIX C

Double-Layer Calculations on Interparticle Repulsion in Clays

Double-layer theory is treated in several text-books (VERWEY and OVERBEEK, 1948; VAN OLPHEN, 1963). The theory is presented here in its simplest form ignoring (a) secondary energy terms and related effects (BOLT, 1955); (b) adsorbed water effects (LOW, 1961; RAVINA and ZASLAVSKY, 1972); and (c) the Stern layer and the effect of ion size (STERN, 1924).

The starting point of the theoretical development is the Poisson equation, which describes the effect of space charge in an electric field on the rate of change of the electrical potential gradient. The clay platelet is regarded as a negatively charged capacitor plate and the neighbouring ions as point charges. Conventionally, in elementary electrostatics theory, 4π lines of force are assumed to emanate from each positive electrostatic unit of charge (esu) and terminate on each negative esu. The electric field intensity (force acting on each negative esu) is assumed to be the number of lines of force per square centimetre in a vacuum. Consider an infinitesimal, rectangular parallelepiped in the electrical field of the clay platelet. The parallelepiped has faces of unit area separated by a distance dx in the direction of the field. Because the parallelepiped includes an excess of positive ions, more lines of force leave its left-hand face than enter its right-hand face, and the difference amounts to $4\pi\rho\ dx$, where ρ is the net space charge density. This difference must equal the change in electrical field intensity, ξ, across the parallelepiped; therefore

$$\frac{d\xi}{dx}\,dx = 4\pi\rho\ dx \qquad \text{(C1)}$$

However,

$$\xi = -\frac{d\psi}{dx} \qquad \text{(C2)}$$

where ψ is the electrical potential. Consequently

$$-\frac{d\xi}{dx} = \frac{d^2\psi}{dx^2} = -4\pi\rho \qquad \text{(C3)}$$

If, instead of a vacuum, the clay platelet and ions are in a medium having a dielectric constant, D, then (C3) becomes

$$\frac{d^2\psi}{dx^2} = -\frac{4\pi\rho}{D} \qquad \text{(C4)}$$

which is the Poisson equation.

In its general form the Boltzmann equation may be written as

$$\frac{n}{n_0} = \exp\left[-\frac{(E - E_0)}{kT}\right] \qquad \text{(C5)}$$

where n is the concentation of particles at a point in a force field where their potential energy is E, n_0 is the concentration of the same particles at a reference point where their potential energy is E_0, k is Boltzmann's constant and T is the absolute temperature. When applied to the distribution of ions in the electric field of a clay platelet

$$E - E_0 = \nu e \psi \qquad \text{(C6)}$$

where ν is the ionic valence and **e** is the elementary charge. Hence (C5) becomes

$$\frac{n}{n_0} = \exp\left[-\frac{\nu e \psi}{kT}\right] \qquad \text{(C7)}$$

The net space charge density, ρ, is given by

$$\rho = e \sum \nu_i n_i \qquad \text{(C8)}$$

where the subscript i signifies any ionic species.

Combining (C4) and (C7) gives the Poisson-Boltzmann equation

$$\frac{d^2\psi}{dx^2} = -\frac{4\pi e}{D}\sum \nu_i\, n_{i0} \exp\left[-\frac{\nu_i e \psi}{kT}\right] \qquad \text{(C9)}$$

If a single symmetrical electrolyte is present, the summation in (C9) includes only two terms and the equation can be written in the following hyperbolic form

$$\frac{d^2\psi}{dx^2} = \frac{8\pi e \nu n_0}{D} \sinh \frac{\nu e \psi}{kT} \qquad \text{(C10)}$$

Making the following substitutions

$$y = \frac{\nu e \psi}{kT} \qquad \text{(C11)}$$

$$z = \frac{\nu e \psi_s}{kT} \qquad \text{(C12)}$$

$$\xi = \chi x \qquad \text{(C13)}$$

$$\chi^2 = \frac{8\pi n_0 e^2 \nu^2}{DkT} \qquad \text{(C14)}$$

where ψ_s is the surface potential, we obtain the following

$$\frac{d^2 y}{d\xi^2} = \sinh y \qquad \text{(C15)}$$

The potential midway between two parallel clay platelets is ψ_m and

$$\mathbf{u} = \frac{\nu\, e\, \psi_m}{k\,T} \tag{C16}$$

Integrating (C15) once with the following boundary conditions

$$\mathbf{x} = d \qquad \mathbf{y} = \mathbf{u} \qquad \text{and} \qquad \frac{d\,\mathbf{y}}{d\,\xi} = 0$$

gives

$$\frac{d\,\mathbf{y}}{d\,\xi} = -\,(2\cosh \mathbf{y} - 2\cosh \mathbf{u})^{0.5} \tag{C17}$$

The second integration between the limits (**z** and **u** for **y**) and (0 and d for **x**) gives

$$\int_{\mathbf{z}}^{\mathbf{u}} (2\cosh \mathbf{y} - 2\cosh \mathbf{u})^{-0.5} = -\int_0^d d\,\xi = -\,\chi\,\mathbf{x} \tag{C18}$$

The charge of the clay platelet is the result of cation substitutions in the crystal and is constant. The potential midway between the clay platelets **u** is a function of the surface potential **z** and the half-distance d and **z** is a function of the surface charge σ and d.

The surface charge σ is given by

$$\sigma = -\int_0^d \rho\, d\,\mathbf{x} \tag{C19}$$

Using (C4)

$$\sigma = -\frac{D}{4\,\pi}\left[\frac{d\,\psi}{d\,\mathbf{x}}\right]_0 \tag{C20}$$

Using (C11) and (C13)

$$\sigma = -\left[\frac{D n_0 kT}{2\pi}\right]^{0.5} \left[\frac{d\,\mathbf{y}}{d\,\xi}\right]_0 \qquad \text{(C21)}$$

Combining (C21) and (C17), recognising that $\mathbf{y} = \mathbf{z}$ when $\xi = 0$, gives

$$\sigma = \left[\frac{D n_0 kT}{2\pi}\right]^{0.5} \left[2\cosh \mathbf{z} - 2\cosh \mathbf{u}\right]^{0.5} \qquad \text{(C22)}$$

Therefore, for constant charge,

$$\left[2\cosh \mathbf{z} - 2\cosh \mathbf{u}\right]^{0.5} \left[n_0\right]^{0.5} = \text{constant} \qquad \text{(C23)}$$

and is independent of the distance between the clay platelets.

To calculate the change of midway potential **u** with varying inter-platelet distance 2 d, at a given external electrolyte concentration n_0, values of **z** and **u** are required whereby

$$\left[2\cosh \mathbf{z} - 2\cosh \mathbf{u}\right]^{0.5} = \sigma\left[\frac{2\pi}{D n_0 kT}\right]^{0.5} = \text{constant} \qquad \text{(C24)}$$

The repulsive force per unit area can be computed as the difference in osmotic pressure midway between clay platelets relative to the external equilibrium solution (LANGMUIR, 1936). The osmotic pressure difference depends directly on the difference in the number of ions in the two regions. For a single cation and anion species of the same valence

$$P \propto (n_m^+ + n_m^-) - (n_0^+ - n_0^-) \qquad \text{(C25)}$$

where the subscripts m and 0 denote the midplane and the external equilibrium solution,

respectively. For the equilibrium solution $n_0^+ = n_0^- = n_0$, where n_0 is the concentration of the external solution. From Boltzmann's equation

$$P \propto n_0 \exp(\mathbf{u}) - n_0 + n_0 \exp(-\mathbf{u}) - n_0 \qquad \text{(C26)}$$

which can be expessed as

$$P \propto 2\, n_0 (\cosh \mathbf{u} - 1) \qquad \text{(C27)}$$

The resulting equation is (van Olphen, 1963)

$$P = 2\, n_0\, k\, T (\cosh \mathbf{u} - 1) \qquad \text{(C28)}$$

Calculation Procedure for the Opalinus Clay

We follow the procedure of MADSEN and MULLER-VONMOOS (1985). The mean value of the specific surface charge, σ (esu.cm^{-2}), is calculated for the clay using

$$\sigma = \frac{\mathbf{e}\, N_A\, C}{S} \cdot 10^{-9} \qquad \text{(C25)}$$

where **e** is the elementary charge (4.80 x 10^{-10} esu), N_A is Avogadro's number (6.02 x 10^{23} ions. g^{-1}), C is the cation exchange capacity of the clay (meq.100g^{-1}) and S is the specific surface area of the clay fraction (m^2.g^{-1}).

Parameter χ is then calculated using

$$\chi = \left[\frac{8 \pi n_0 e^2 v^2}{D k T} \right]^{0.5} \qquad \text{(C30)}$$

where **e** is the elementary charge (4.80 x 10^{-10} esu), D is the dielectric constant of water (80) and k is Boltzmann's constant (1.38 x 10^{-16} erg.K^{-1}). The ionic concentration of the macropore water n_0 is then given by $C_p \cdot N_A \cdot 10^{-3}$ where C_p is the macropore

solute concentration (mol.l^{-1}).

The rate of change of electrical potential with respect to ξ at the surface of the clay platelet is found using

$$\left[\frac{d\,y}{d\,\xi}\right]_0 = \frac{4\,\pi\,e\,\nu\,\sigma}{D\,k\,T\,\chi} \qquad \text{(C31)}$$

Values of z are then determined for multiple values of **u** using

$$\cosh \mathbf{z} = 0.5\left[\frac{d\,y}{d\,\xi}\right]_0^2 + \cosh \mathbf{u} \qquad \text{(C32)}$$

and the product χ d is found for each value of **u** by integrating

$$\chi\ d = -\int_{z}^{u} (2\cosh \mathbf{y} - 2\cosh \mathbf{u})^{-0.5}\, d\mathbf{y} \qquad \text{(C33)}$$

For the puposes of our scoping calculations, approximate solutions of (C33) were obtained using a numerical procedure, enabling the half-distance d between clay platelets to be determined. The pressure P (MPa) is then calculated for each value of **u** using

$$P = 2\,n_0\,k\,T\,(\cosh \mathbf{u} - 1)\ \cdot 10^{-7} \qquad \text{(C34)}$$

Since P is defined as the osmotic pressure relative to the external (macropore) solution, we can equate it directly with the R_{diff} term of Section 6.4.1.