EFFECT OF ALTERATION ON PORE STRUCTURE OF CRYSSTALLINE ROCKS:
CORE SAMPLES FROM ATIKOKAN, ONTARIO

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ABSTRACT

Recent studies indicate that the extensive micropore net-
work within the unfractured rock around major fractures
may have a significant effect in reducing the rate of ra-
dionuclide migration. The micropore structure of granite
samples from Atikokan, Ontario has been studied by
analyzing the results of porosity, electrical and permeability
measurements. The effective porosity, connecting porosi-
ty, tortuosity and permeability are calculated from these
measurements; their values for these samples are in the order
of 0.19-0.64%, 0.06-0.34%, 1.7-4.5 and 0.1-54.9 micro-
darcies, respectively. Results indicate that alteration reduces
the connecting porosity and permeability, and increases the
tortuosity. This is explained by reduction of the aperture
of pore pathways due to dissolution of certain minerals
followed by deposition of secondary minerals. The results
also suggest that laboratory data for permeability may be
extended to field conditions below a certain depth (about
400 metres).

Keywords: pore structure, connecting porosity, effective
porosity, tortuosity, permeability, alteration effects, ra-
dionuclide migration, granite, Atikokan (Ontario),
nuclear-fuel waste.

INTRODUCTION

The Canadian Nuclear Fuel Waste Management
program is assessing the concept that high-level nuclear-fuel waste can be isolated from the biosphere
by means of deep, subsurface disposal in stable geological formations such as granitic plutons in the
Canadian Shield (Boulton 1978, Shemilt 1982).

Though the rock mass around a disposal vault would
provide a natural barrier between the waste and the
biosphere, the potential for contamination of
groundwater by radionuclides dissolved from the
waste and its eventual migration into the biosphere
is a major long-term concern (Shemilt 1982, Scott
1979). Although it is generally accepted that the most
probable path of radionuclide migration would be
along systems of major fractures, recent studies (e.g.,
Neretnieks 1980, Grisak & Pickens 1981, Katsube
1982) point out the effect that unfractured rock
might have on retarding the rate of migration and
storing the radionuclides. The pore structure of the
unfractured rock would control these effects.

Though crystalline rocks like granite may be refer-
ted to as a single homogeneous mass in terms of rock
type, heterogeneities in modal composition, grain
size, degree of fracturing and of alteration may be
present throughout the body, either on local or
regional scales. This is well documented in the case
of the Eye-Dashwa Lakes granitic pluton near
Atikokan, Ontario; alteration in this intrusive body
is concentrated in the vicinity of fracture zones
(Kamineni & Dugal 1982). Rock alteration may af-
fect the pore structure. Porosity in crystalline rocks
rocks can be mainly of two types: 1) primary, acquired dur-
ing crystallization and 2) secondary, acquired since

crystallization through rock alteration. Porosity may
also be a result of expansion by unloading. These
variations may have an effect on radionuclide migra-
tion and storage rates within a rock mass. This paper
focuses mainly on the influence of rock alteration
on pore structure; it is based on the results obtained
from studies made on core samples from Atikokan,
Ontario.

GEOLOGICAL CHARACTERISTICS OF THE ROCK MASS

The Atikokan site (see Fig. 1 in Kamineni et al.
1983, this issue) is one of the areas being investigated
by the Atomic Energy of Canada Limited for generic
research related to the disposal of nuclear-fuel waste in crystalline rocks. The research area is confined to the Eye-Dashwa granitic pluton, located about 30 km northeast of the town of Atikokan. The pluton occurs in the Superior structural province of the Canadian Shield.

The Eye-Dashwa pluton is an egg-shaped body (Brown et al. 1980) with a granodioritic rim and a granitic core (Kamineni & Brown 1981). The lithological differences between the core and the rim are reflected by mineral zonation in the pluton. hornblende is concentrated near the rim, whereas biotite is the predominant mafic mineral in the core. Detailed geological and hydrogeological investigations were undertaken on the southeastern rim of the pluton. Five boreholes (ATK 1, 2, 3, 4 and 5) were drilled; ATK 1 and 5 exceed 1000 m in length, whereas the others are less than 400 m in depth.

Core samples recovered from boreholes display various degrees of alteration (Kamineni & Dugal 1982). Rock alteration can be recognized by both physical and chemical signatures. Altered rock is characterized by a pink color, the intensity of which varies with the degree of alteration, ranging from deep pink in the highly altered regions through lighter shades to grey in the unaltered areas. Under the microscope, the altered rocks contain various secondary minerals such as epidote, sericite, chlorite, carbonate and hematite. These alteration products were developed from primary minerals mostly during the late magmatic to hydrothermal stage (Kamineni & Dugal 1982). The minerals most affected by alteration include hornblende, biotite and plagioclase.

Compared to the unaltered rocks, the altered rocks show 1) depletion of Ca and Sr, 2) slight enrichment of Mg, and 3) higher oxidation ratios of Fe (Kamineni & Dugal 1982).

Pore Structure

Unfractured crystalline igneous rocks consist of mineral grains that cover a wide range in size. Open microfractures may develop along these grain boundaries, within these grains and across grains (Simmons & Richter 1976). Brace (1977), Montgomery & Brace (1975), Sprunt & Brace (1974), Keller (in Katsube & Collett 1973, Parkhomenko 1967), and Norton & Knapp (1977) have provided a considerable amount of basic information on the structure of the pores along these microfractures. Based on this information and on studies of granite specimens using the scanning-electron microscope (Chernis 1981, Katsube et al. 1982a,b), a number of pore-structure models have been developed (Katsube 1981).

Pores in a crystalline rock can belong to one of three types (Fig. 1) based on these models: connecting pores, pocket pores and isolated pores. Connecting pores are generally continuous passages that traverse a rock, and contribute to the migration of radionuclides. Pocket pores are the pocket-shaped enlargements along the connecting pores, or the dead-end pores that branch off from the connecting pores. These pores contribute to the increase of porosity but do not necessarily contribute to the migration. On the contrary, they probably will retard the migration of radionuclides (Katsube 1982, Neretnieks 1980, Grisak & Pickens 1981, Taw et al. 1980). Isolated pores are equant or vug-like pores that appear to be isolated within mineral grains.

Connecting porosity $\Phi_c$, effective porosity $\Phi_e$ and tortuosity $\tau$ are the three principal pore-parameters that are known to affect radionuclide migration (Katsube 1982). Connecting porosity refers to the porosity of the connecting pores, with the assumption that they are parallel to the general direction of flow. If it is assumed that the general direction of flow and pore structure are as shown in Figure 2, these parameters can be defined as follows:

$$\Phi_c = \frac{(Ad \times \ell)\ell}{\ell^3} = \frac{Ad}{\ell}$$  (1)

$$\Phi_e = \Phi_c + \frac{\ell(\ell_p - \ell)Ad}{\ell^3} + \Phi_p = \frac{\ell_p}{\ell} \Phi_c + \Phi_p$$  (2)

$$\tau = \frac{\Phi_e}{\Phi_c} = \frac{\ell_p}{\ell} + \frac{\Phi_p}{\Phi_c}$$  (3)

where $Ad$ is the accumulated aperture of all pores and fractures (m), $\ell$ is the length of one side of a cube of rock under consideration (Fig. 2) (m), $\ell_p$ is the actual length of the tortuous pore or fracture (m),
and $\Phi_p$ is the porosity of pocket pores. The concept of $Ad$ is based on the assumption that all the pores and fractures are parallel, continuous sheet-like openings, but with different apertures. Effective porosity $\Phi_E$ is the sum of connecting porosity $\Phi_c$, the excess porosity caused by a connecting pore being tortuous, and the porosity of the pocket pores $\Phi_p$. If $\Phi_p$ is equal to 0 and the connecting pores are not tortuous, $\ell_p$ is equal to $\ell$, then $\Phi_c$ and $\Phi_p$ are equivalent, and $\tau$ is equal to 1. The method of introducing these parameters into the diffusion, advection and diffusion-advection equations is described in detail by Wadden & Katsube (1982) and by Katsube (1982). Pocket pores (Fig. 1) will contribute to storage.

The effective porosity $\Phi_E$ is determined by measuring the difference in mass between the rock samples when vacuum-saturated and oven-dried. The connecting porosity $\Phi_c$ is derived from $\Phi_E$ and the formation factor $F$. The formation factor was introduced by Archie (1942) and is defined by,

$$ F = \frac{\rho_p}{\rho_{pw}} $$

and

$$ \Phi_c = \sqrt{\frac{\Phi_E - \Phi_p}{F}} $$

where $\rho_p$ is the electrical resistivity of a water-saturated rock, and $\rho_{pw}$ is the electrical resistivity of a pore water. The actual method used to measure the values of $\Phi_E$ and $F$ in this study is described in Katsube (1981). According to the pore model described by Ward & Fraser (1967),

$$ F = \frac{\tau'}{(Ad/\ell)} $$

where $\tau'$, the true tortuosity, is defined as

$$ \tau' = \frac{\ell_p}{\ell} $$

From equations 1, 2, 5 and 6, the connecting porosity can be expressed by

$$ \Phi_c = \sqrt{\frac{\Phi_E - \Phi_p}{F}} $$
Permeability $k$ is not a basic pore-parameter, but is dependent on $\tau$ and $\Theta_c$ (Katsube 1982); it emphasizes the effect of the individual apertures $d$:

$$k = \frac{d^2 \Theta_c}{12\tau} = \frac{d^2 \Theta_e}{12\tau^2}$$

(8)

where $d$ represents the fracture aperture of pores.

Tortuosity $\tau$ rather than true tortuosity $\tau'$ is used here because the effect that pocket pores have on permeability $k$ is not clearly understood.

SAMPLES AND METHOD OF INVESTIGATION

Thirty-five samples of granite were taken at intervals ranging from 10 to 50 metres along the core of borehole ATK-I in the Atikokan research area. The length of this borehole is 1150 metres. It cuts through granite showing various degrees of alteration. Each sample 70 centimetres in length was cut into seven 10-cm specimens, and then distributed to different investigators. The specimen labeled "P" was then cut into seven subspecimens (Fig. 3). Various tests were performed on each specimen. The results used in this study were obtained from specimens b.1 (immersion porosity), c (electrical resistivity) and e (permeability).

The samples investigated were divided into various groups according to their degree of alteration. The degree of alteration can be determined 1) qualitatively, by the intensity of the pink color, and 2) quantitatively, through microscopic examination of the amount of secondary minerals present. The least altered rocks are grey and show no trace of pink color. Few or no secondary minerals are present in these rocks. The altered rocks are pink and contain abundant secondary minerals. In other words, the intensity of pink color and the modal proportion of secondary minerals are positively correlated.

Among the samples investigated, four groups were distinguished according to the intensity of pink coloration: 1) grey, 2) pinkish grey, 3) grey pink, and 4) pink. Petrographically, the primary minerals in grey samples show the least alteration; the plagioclase is altered around its rim to sericite and kaolin, whereas biotite and hornblende are slightly altered to chlorite and epidote. Compared to the grey samples, the samples belonging to groups 2 and 3 (pinkish grey and grey pink) show much higher amounts of the secondary minerals epidote, chlorite, sericite, carbonate and kaolin. There is a corresponding decrease in the amounts of primary minerals. The pink-colored samples (group 4), which are the most altered, show complete conversion of hornblende and biotite to epidote and chlorite. Some titanite, carbonate and quartz have also formed as secondary products. Plagioclase grains in these rocks are converted extensively, more commonly to sericite and epidote and less commonly to carbonate and chlorite. The general distribution of these four groups of rocks is represented as zones along the core from the ATK-I borehole (Fig. 4).

The effective porosity $\Theta_e$ and formation factor $F$ are measured for all samples by methods described previously. Then the values of $\Theta_e$ are derived from

Fig. 3. Location of specimens within the sample and description of type of test performed on them.
The parameter $\phi_o$ decreases and $\tau$ increases with increasing intensity of alteration (Fig. 4). A trend similar to $\phi_o$ is seen for $\phi_E$, but to a less obvious degree (Fig. 4). Both permeabilities ($k_o$, $k_e$) decrease with increasing degree of alteration, and are generally high (6-20 microdarcies) for unaltered rocks. A fracture was observed in sample ATK-I-110, from a depth of 110 metres. This is probably the cause of the high value of its permeability.

The values for the five parameters are listed in Table 1. These values are generally in the same range as those for the granite samples from the Whiteshell research area, Piwawa, Manitoba (Katsube 1981). The permeability values are in the intermediate range for granites, in terms of the results reported by Brace (1980), which are in the range $10^{-3}$–30 microdarcies. Tension fractures due to stress release develop in rock samples that are taken at depth (Annor & Katsube 1983). The effect of stress release is reflected in the values of $k_e$ (Fig. 5), but not in those of $k_o$. However, it is interesting that the effect of stress release is much smaller on altered rocks than on unaltered rocks. Further study of this subject is under way.

### Results

Connecting porosity $\phi_o$, tortuosity $\tau$, effective porosity $\phi_E$, permeabilities for unconfined stress $k_o$ and for confined stress (equivalent to overburden stress) $k_e$ and intensity of alteration were determined for 35 samples from borehole ATK–1. The samples range in depth from 13 to 1140 metres. The results are shown in Figure 4 for $\phi_o$, $\tau$ and $\phi_E$, and in Figure 5 for $k_o$ and $k_e$.

The parameter $\phi_o$ decreases and $\tau$ increases with increasing intensity of alteration (Fig. 4). A trend similar to $\phi_o$ is seen for $\phi_E$, but to a less obvious degree (Fig. 4). Both permeabilities ($k_o$, $k_e$) decrease with increasing degree of alteration, and are generally high (6-20 microdarcies) for unaltered rocks. A fracture was observed in sample ATK–1–110, from a depth of 110 metres. This is probably the cause of the high value of its permeability.

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### Table 1: Pore Characteristics and Degree of Alteration of Atikokan Specimens

<table>
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<th>Sample/Depth</th>
<th>$\phi_o$</th>
<th>$\phi_E$</th>
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<th>$k_o$</th>
<th>$k_e$</th>
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**Notes:**
- $\phi_o$: effective porosity.
- $\phi_E$: connecting porosity.
- $\tau$: tortuosity.
- $k_o$: permeability at unconfined pressure of 1.4 MPa.
- $k_e$: permeability at confined pressure equal to overburden pressure.
The permeabilities $k_o$, $k_c$ show a good relationship with alteration (Fig. 5). All samples with alteration intensities 3 and 4 show permeabilities below 1.4 microdarcies, and 93% of those with alteration intensities of 1 and 2 show values above 1.4 microdarcies. The $k_o$ vs. $\Omega_E$ plots are shown in Figure 7. The correlation is moderately good ($r$ (correlation coefficient) 0.74, $n$ (gradient) 4.4) but, unlike the cases of $r$ vs. $\Omega_E$, strong characteristics of a single modal distribution are seen if the plots in Figure 7 are projected on the line A–B (mean -0.18, standard deviation 0.18).

Results shown in Figure 6 indicate that tortuosity values fall into either one of two populations, above or below 2.5. A higher value of tortuosity implies either a path with higher tortuosity, or a higher ratio of porosity in the pocket and blind pores over that in the connecting pores (Katsube 1982, Wadden & Katsube 1982), as shown in Figure 8. Since it is assumed that $\Omega_p$ is 0 when deriving $\Omega_p$ from equation 7, the actual value of $\tau$ could be larger than that derived from equation 3 ($\Omega_p$ is not necessarily equal to zero in equation 7). Kamineni & Dugal (1982) and others (e.g., Ferry 1979) have suggested that a volume increase of secondary minerals takes place in pores as alteration progresses. Since dissolution and deposition of certain minerals would accompany the alteration, intuitively the increase in pocket porosity $\Omega_p$ over connecting porosity (Fig. 8) is a realistic model. It implies that the aperture of the connecting pores would decrease with alteration. This would occur if a rise in temperature were followed by a fall in temperature, which would cause dissolution of certain minerals and increase the pocket porosity, followed by supersaturation of chemical constituents and deposition along all pores including connecting pores. Permeability $k$ is sensitive to change in aperture, as is obvious from equation 8. If $k_o$ were dependent only on $\Omega_E$, then the $k_o$ vs. $\Omega_E$ should have a slope of 1 on the log-log scale. The fact that the true slope (Fig. 7) is closer to 4 suggests that the aperture $d$ of the connecting pores increases with increasing values of $k_o$ ($r$ can be considered constant). These data, therefore, support the alteration model in Figure 8.

Ferry (1979) noted that the alteration of primary minerals to epidote, chlorite, carbonate and muscovite in granitic rocks from south-central Maine is accompanied by an increase in volume. Fracture-controlled alteration, leading to development of smectite, zeolites and celadonite and healing of fractures, has been reported in submarine basalts (Thompson 1973, Fyfe 1974). Self-sealing of pores and microfractures has also been noted in geothermal areas, presumably due to increase in volume (Browne 1978). The experiments of Summers et al. (1978) on the Westerly granite provide further infor-

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**Discussion**

The existence of a relationship between tortuosity $\tau$ and alteration (Fig. 4) is significant. The relationship is more distinct in Figure 6. The values for effective porosity $\Omega_p$ vary between 0.19 and 0.64%, and this range of variation is generally not affected by alteration. However, all the rock samples with the higher intensities of alteration (groups 3 and 4) show tortuosity values larger than 2.5, and 88% of those with the lower intensities of alteration (groups 1 and 2) show tortuosity values below 2.5 ($\tau$ vs. $\Omega_E$ plots and frequency-distribution diagram in Fig. 6).
mation on alteration of matrix minerals and its influence on permeability. Summers et al. studied water flow through core samples for periods of time up to 17 days at temperatures of 100°, 200°, 300° and 400°C. In all cases, the initial permeability was from 1 to 2 orders of magnitude higher at elevated temperatures than at room temperature, perhaps owing to thermal stress cracking, but in nearly all cases the permeability decreased significantly during the test period. Dissolution of plagioclase and quartz occurred near the inlet, where the pore pressures are the highest. Precipitation occurred throughout the samples because of oversaturation as the pore pressure dropped, causing significant reduction in permeability.

Preliminary comparison of permeability values with rock-mass permeabilities measured in borehole ATK-1 by Gartner Lee and Associates (1982) shows some interesting trends. Three zones of rock-mass permeabilities were noted in the borehole. Above 250 m, the permeability ranges from $10^2$ to $10^4$ microdarcies, between 250 and 400 m it ranges from $10^{-1}$ to 10 microdarcies, and below 400 m the values are less than $10^{-1}$ microdarcies. The values noted in the deep zone are similar to the matrix-permeability values reported here. The high values of rock-mass permeability noted in the shallow part of the borehole, in contrast to matrix-permeability values, indicate that the permeability in this altered zone is controlled by fractures, whereas at deeper levels, fractures play an insignificant role. The fractures in the upper zones probably have larger apertures owing to erosional unloading of cover rocks or melting of Pleistocene glaciers. From this, it is suggested that rock-mass permeability and matrix permeability show diverging trends at shallow depths and converging trends at deeper level. This implies that the laboratory results for permeability may represent the in situ conditions.

**Conclusions**

The pore structure and alteration data contained in this paper suggest that alteration affects the pore structure; the effect is reflected in a number of pore parameters. Tortuosity $\tau$ and permeability $k$ are probably the two parameters that reflect this effect to the greatest extent. These parameters have a significant effect on radionuclide migration (Katsube 1982, Wadden & Katsube 1982).
The effect of alteration on pore structure is explained by the dissolution of certain minerals at high temperatures, causing an increase in pocket porosity, followed by deposition of secondary minerals due to supersaturation at lower temperatures; these minerals reduce the aperture of the pore pathways. There are a number of previous studies that support this concept. Preliminary inspection of in situ and laboratory data on permeability at depth, where, presumably, expansion of fracture aperture due to erosional unloading of cover rocks or melting of Pleistocene glaciers is not significant, suggest that conditions drawn on the basis of laboratory data may be extended to field conditions for depth greater than 400 metres.

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