Canadian Mineralogist
Vol. 21, pp. 637-646 (1983)

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

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ABSTRACT

Recent studies indicate that the extensive micropore network within the unfractured rock around major fractures may have a significant effect in reducing the rate of radionuclide 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 porosity, 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 microdarcies, 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, radionuclide migration, granite, Atikokan (Ontario), nuclear-fuel waste.

SOMMAIRE

Des études récentes montrent qu'un réseau suffisamment vaste de micropores dans la roche non-fracturée qui avoisine les fractures principales pourrait retarder de façon marquée le taux de migration des radionucléides. La structure des micropores d'échantillons de granite provenant d'Atikokan (Ontario) est étudiée par analyse de porosité et mesures de propriétés électriques et de perméabilité. On déduit de ces mesures la porosité effective, la porosité des liaisons, la tortuosité et la perméabilité. Pour ces échantillons, on trouve les valeurs suivantes: 0.19–0.64%, 0.60–0.34%, 1.7–4.5 et 0.1–54.9 microdarcies. Ces résultats montrent que, par altération, la porosité et la perméabilité diminuent, tandis que la tortuosité augmente. Ces phénomènes seraient dus à la constriction des pores par dissolution de certains minéraux et dépôt de minéraux secondaires. Les mesures de perméabilité faites au laboratoire resteraient valides sur le terrain à plus de 400 mètres de profondeur.

(Traduit par la Rédaction)

Mots-clés: structure des pores, porosité des liaisons, porosité effective, tortuosité, perméabilité, effets d'altération, migration de radionucléides, granite, Atikokan (Ontario), déchets nucléaires.

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 referred 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 affect the pore structure. Porosity in crystalline rocks can be mainly of two types: 1) primary, acquired during 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 migration 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, Tang *et al.* 1980). Isolated pores are equant or vug-like pores that appear to be isolated within mineral grains.

Connecting porosity $\varnothing_c$, effective porosity $\varnothing_E$ and tortuosity τ 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:

$$\varnothing_c = \frac{(\mathrm{Ad} \times \ell)\ell}{\ell^3} = \frac{\mathrm{Ad}}{\ell} \quad (1)$$

$$\varnothing_E = \varnothing_c + \frac{\ell(\ell_p - \ell)\mathrm{Ad}}{\ell^3} + \varnothing_p = \frac{\ell_p}{\ell}\varnothing_c + \varnothing_p \quad (2)$$

$$\tau = \frac{\varnothing_E}{\varnothing_c} = \frac{\ell_p}{\ell} + \frac{\varnothing_p}{\varnothing_c} \quad (3)$$

where Ad is the accumulated aperture of all pores and fractures (m), ℓ is the length of one side of a cube of rock under consideration (Fig. 2) (m), ℓ_p is the actual length of the tortuous pore or fracture (m),

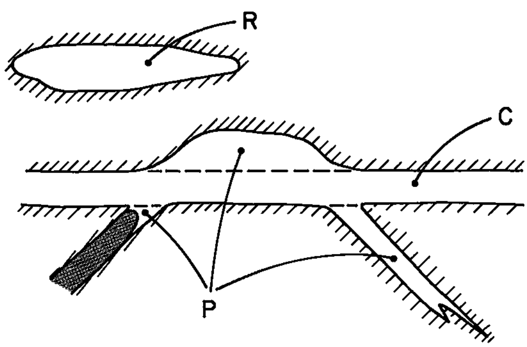

FIG. 1. Basic pore-structure model. Legend: C connecting pores, P pocket pores, R isolated pores.

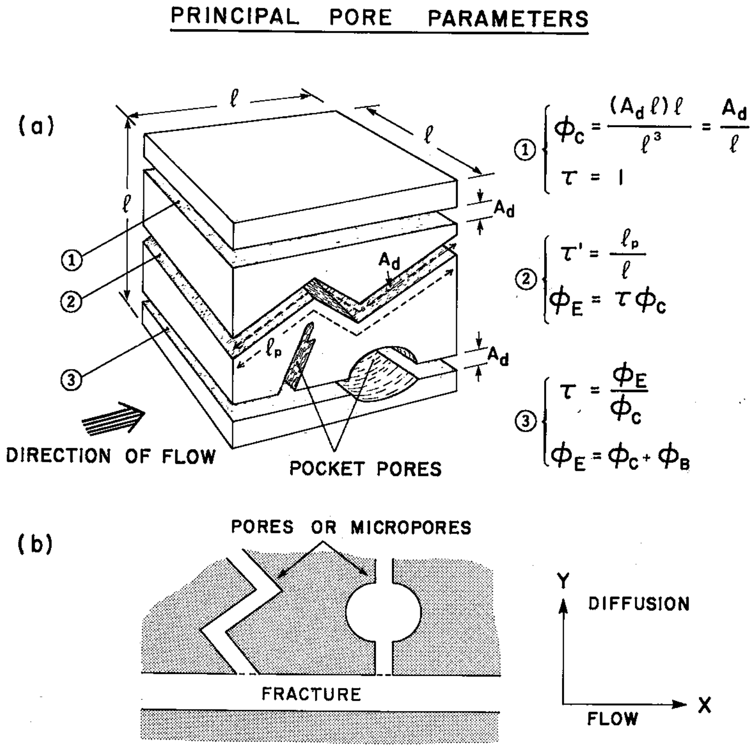

FIG. 2. Pore parameters that influence radionuclide migration (after Katsube 1982). (1) Nontortuous connecting pore, (2) tortuous connecting pore, and (3) pocket pores along a nontortuous connecting pore.

and $\varnothing_p$ 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 $\varnothing_E$ is the sum of connecting porosity $\varnothing_c$, the excess porosity caused by a connecting pore being tortuous, and the porosity of the pocket pores $\varnothing_p$. If $\varnothing_p$ is equal to 0 and the connecting pores are not tortuous, ℓ_p is equal to ℓ, then $\varnothing_E$ and $\varnothing_c$ are equivalent, and τ 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 $\varnothing_E$ is determined by measuring the difference in mass between the rock samples when vacuum-saturated and oven-dried. The connecting porosity $\varnothing_c$ is derived from $\varnothing_E$ and the formation factor F. The formation factor was introduced by Archie (1942) and is defined by,

$$F = \frac{\rho_p}{\rho_{pw}}, \tag{4}$$

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

$$F = \frac{\tau'}{(\mathrm{Ad}/\ell)} \tag{5}$$

where τ', the true tortuosity, is defined as

$$\tau' = \frac{\ell_p}{\ell} \tag{6}$$

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

$$\varnothing_c = \sqrt{\frac{\varnothing_E - \varnothing_p}{F}} \tag{7}$$

Permeability k is not a basic pore-parameter, but is dependent on τ and $\emptyset_c$ (Katsube 1982); it emphasizes the effect of the individual apertures d:

$$k = \frac{d^2}{12\tau} \emptyset_c = \frac{d^2}{12\tau^2} \emptyset_E \tag{8}$$

where d represents the fracture aperture of pores.

Tortuosity τ rather than true tortuosity τ' 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-1 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-1 borehole (Fig. 4).

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

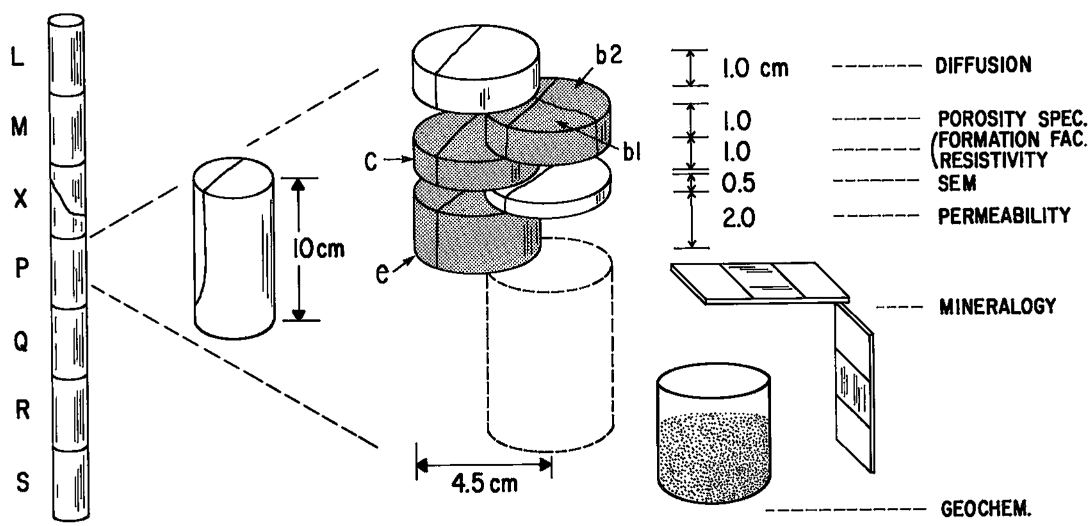

Fig. 3. Location of specimens within the sample and description of type of test performed on them.

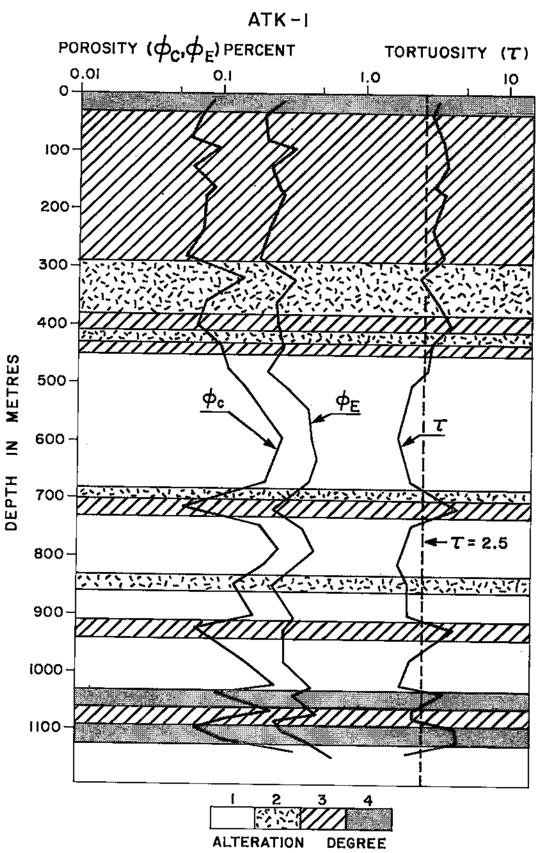

FIG. 4. Variation in connecting porosity $\emptyset_c$, effective porosity $\emptyset_E$ and tortuosity τ with depth (borehole ATK-1) and degree of alteration in the Atikokan research area.

equation (7) using these results, assuming that $\emptyset_p$ is 0 for convenience. Tortuosity τ is derived from equation (3). The permeability is determined at two confining pressures: 1.4 MPa and equivalent overburden pressure. The two permeabilities are expressed by k_o and k_c, respectively. The basic principles used to make these measurements are described in the paper by Brace *et al.* (1968), and the methods used to apply these principles to this project are discussed in Katsube (1981). The error range for $\emptyset_E$ and F is in the order of ±10 and ±20–40 %, respectively. The resolution of the permeability measurements is in the order of 0.05 microdarcies.

RESULTS

Connecting porosity $\emptyset_c$, tortuosity τ, effective porosity $\emptyset_E$, permeabilities for unconfined stress k_o and for confined stress (equivalent to overburden stress) k_c 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 $\emptyset_c$, τ and $\emptyset_E$, and in Figure 5 for k_o and k_c.

The parameter $\emptyset_c$ decreases and τ increases with increasing intensity of alteration (Fig. 4). A trend similar to $\emptyset_c$ is seen for $\emptyset_E$, but to a less obvious degree (Fig. 4). Both permeabilities (k_o, k_c) 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.

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_o (Fig. 5), but not in those of k_c. 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.

TABLE 1. PORE CHARACTERISTICS AND DEGREE OF ALTERATION OF ATIKOKAN SPECIMENS

Sample/ Depth	$\emptyset_E$	$\emptyset_c$	τ	k_o	k_c	Alteration
ATK-1 - 13	0.27	0.085	3.2	0.1	0.6	4
39	0.20	0.071	2.9	0.7	1.1	3
79	0.21	0.060	3.4	0.3	-	3
94	0.34	0.096	3.5	0.2	0.1	3
110	-	-	-	17.6	6.8	3
128	0.23	0.062	3.7	0.4	0.2	3
163	0.26	0.088	3.0	0.8	0.3	3
174	0.28	0.077	3.6	0.1	0.1	3
237	0.22	0.075	2.9	0.3	0.3	3
283	0.19	0.055	3.5	0.1	0.2	3
319	0.34	0.141	2.4	1.5	0.9	2
361	0.25	0.078	3.2	0.3	0.3	2
400	0.26	0.068	3.9	0.4	0.5	3
433	0.28	0.099	2.9	0.4	0.3	2
475	0.32	0.111	2.8	1.3	0.6	1
505	0.31	0.147	2.1	1.7	0.7	1
539	0.43	-	-	4.5	1.2	1
593	0.45	0.267	1.7	4.0	0.5	1
630	0.49	0.286	1.7	16.6	6.0	1
670	0.43	0.204	2.1	7.4	1.0	1
714	0.24	0.055	4.4	0.3	0.1	3
746	0.39	0.187	2.1	3.8	0.6	2
787	0.47	0.253	1.9	17.1	1.3	1
812	0.35	0.208	1.7	7.8	1.5	1
848	0.24	0.121	2.0	0.5	0.1	1
901	0.34	0.172	2.0	3.0	0.3	1
923	0.29	0.066	4.3	0.3	0.1	3
979	0.29	0.141	2.1	2.1	0.3	1
1021	0.46	0.247	1.8	13.0	1.8	1
1034	0.34	0.095	3.6	0.2	0.1	4
1063	0.49	0.225	2.3	25.1	1.4	1
1078	0.25	0.111	2.2	16.6	-	3
1096	0.29	0.065	4.4	0.1	0.1	4
1120	0.46	0.102	4.5	0.5	0.3	4
1140	0.64	0.344	1.9	54.9	-	1
(m) UNITS	%	%		μ-d	μ-d	

$\emptyset_E$ effective porosity, $\emptyset_c$ connecting porosity, τ tortuosity

k_o permeability at confining pressure of 1.4 MPa

k_c permeability at confining pressure equal to overburden pressure

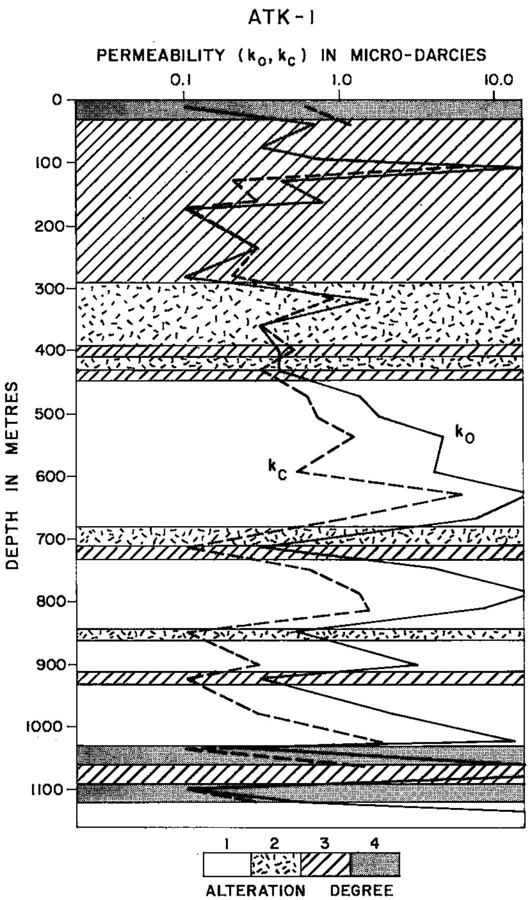

FIG. 5. Variation in hydraulic permeability k with depth and degree of alteration in the Atikokan research area; k_o, k_c are hydraulic permeabilities measured under confining pressures of 1.4 MPa and equivalent overburden stress, respectively (after Katsube 1982).

DISCUSSION

The existence of a relationship between tortuosity τ and alteration (Fig. 4) is significant. The relationship is more distinct in Figure 6. The values for effective porosity $\varnothing_E$ 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 intensitites 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 (τ *vs.* $\varnothing_E$ plots and frequency-distribution diagram in Fig. 6).

The permeabilities k_o, k_c show a good relationship with alteration (Fig. 5). All samples with alteration intensitites 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.* $\varnothing_E$ 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 τ *vs.* $\varnothing_E$, 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 $\varnothing_p$ is 0 when deriving $\varnothing_c$ from equation 7, the actual value of τ could be larger than that derived from equation 3 ($\varnothing_p$ 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 $\varnothing_p$ 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 $\varnothing_E$, then the k_o *vs.* $\varnothing_E$ 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 (τ 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-

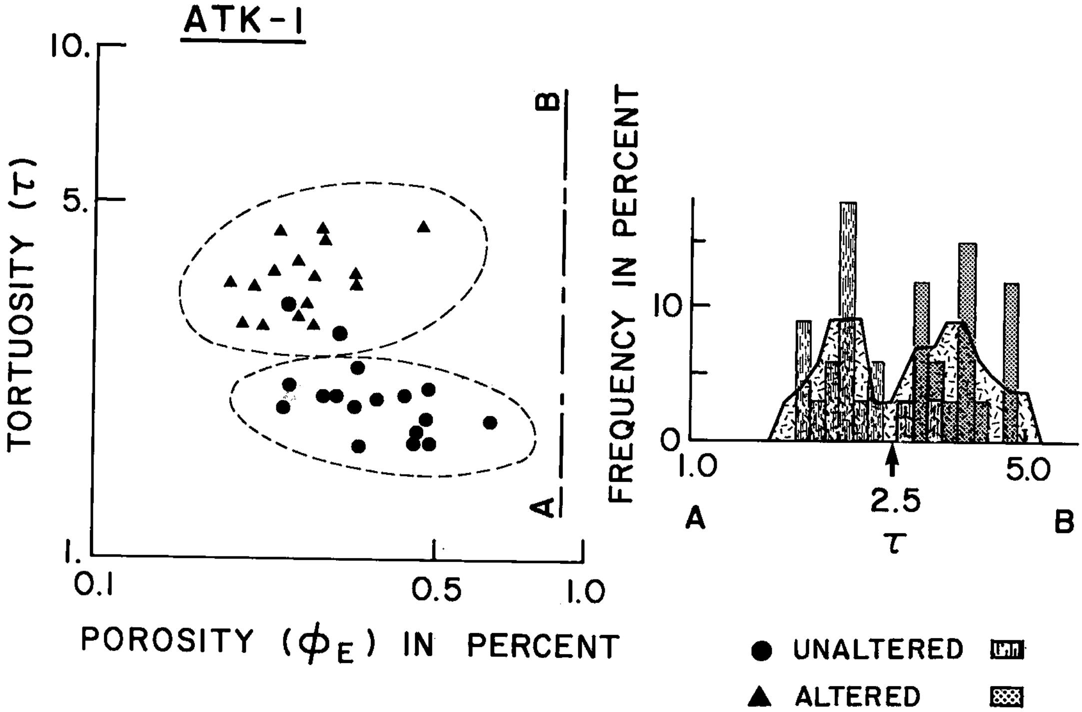

FIG. 6. Relationship between alteration and tortuosity τ (after Katsube 1982). The mean tortuosity $\bar{\tau}$ of unaltered samples is 2.0 (standard deviation σ 0.3), and that of altered samples is 3.6 (σ 0.6).

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 propably 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 τ 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).

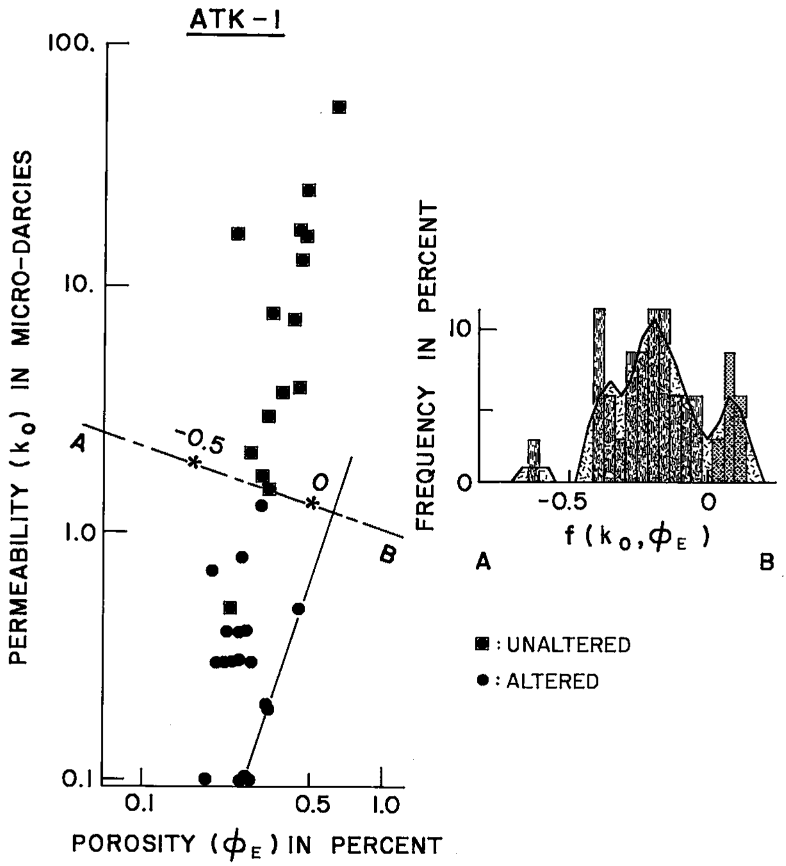

FIG. 7. Relationship between permeability and degree of alteration. (a) Relationship between permeability k_o and effective porosity $\emptyset_E$: r (correlation coefficient) = 0.74, n (gradient) = 4.4. (b) Histogram of plots when projected on line A–B.

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.

ACKNOWLEDGEMENTS

The authors ackowledge the comments and critical review of the manuscript by K.L. Currie and R.W. Boyle (Geological Survey of Canada). The authors also are grateful to N.C. Wardlaw (University of Calgary), an anonymous reviewer, R.F. Martin (McGill University) and J.B. Percival (Atomic Energy of Canada Limited) for their comments. The main part of this work was supported by the Atomic Energy of Canada Limited. Electrical resistivity measurements were carried out at the Electrical Rock Property Laboratory in the Geological Survey of Canada. The permeability measurements were performed at Terratek, Salt Lake City, Utah, U.S.A.

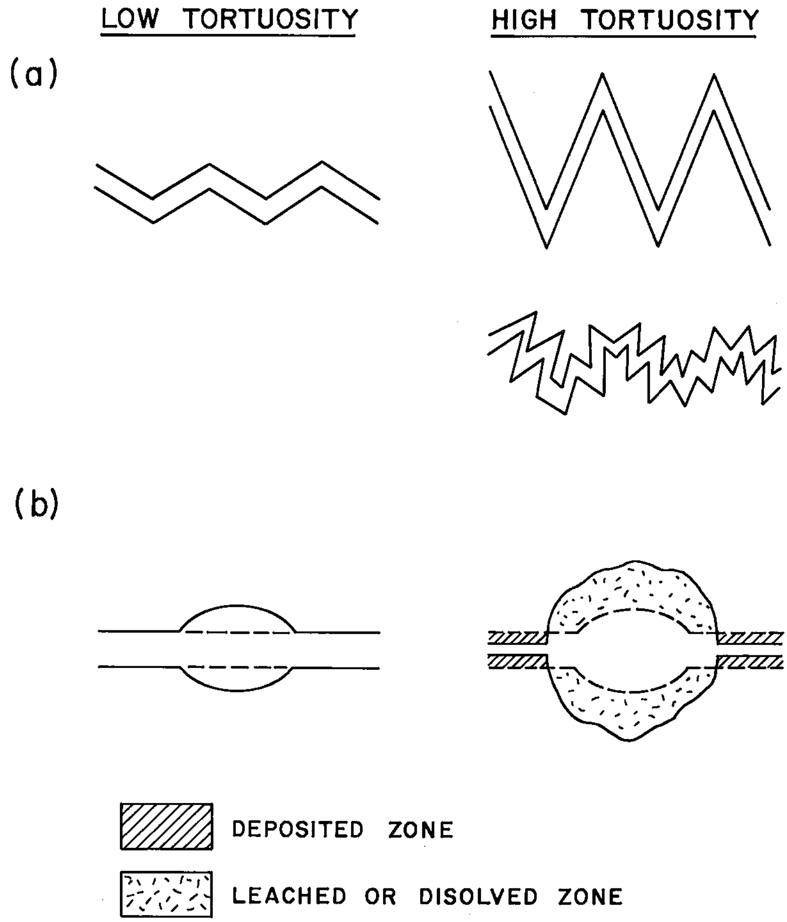

FIG. 8. Implication of tortuosity increase (a), the effect of alteration on pore structure (b).

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*Unrestricted internal report available from SDDO, Atomic Energy of Canada Limited Research Company, Chalk River, Ont. K0J 1J0.

Received January 10, 1983, revised manuscript accepted August 10, 1983.