MINERALOGY AND FABRIC OF FOUR SWELLING SHALES
膨脹性 쉐일들의 鑛物組成과 層狀構造

要 旨

本稿는 膨脹性 쉐일들의 時間依存的變形을 더 잘 理解하기위히 實行한 鑛物組成, 層狀構造 그리고 空隙徑分布에 對한 分析節次와 그 結果에 關하여 說明한다. 쉐일들의 鑛物組成은 X線回折分析과 化學分析結果에 의히 糾明한다. 그리고 쉐일의 膨脹性擧動을 理解하는데 있어서의 層狀構造의 重要性이 走査 電子顯微鏡에 의히 實證된다. 또한 쉐일이 膨脹하는 過程에서 생기는 空隙徑分布의 變化를 自由膨脹 試驗前과 後의 空隙徑分布를 測定하여 調査한다.

硏究結果로 보면 調査한 쉐일들의 膨脹은 黃鐵石, 硬石膏 또는 膨脹性 粘土鑛物들과 같은 鑛物들에 의히 起因하는것이 아님이 糾明되었다. 이 쉐일들의 異方性 膨脹擧動은 各各의 層狀構造와 그들의 差異에 의히 說明할 수 있었다. 쉐일들이 膨脹하는 過程에서 空隙의 體積이 增加하는 것이 밝혀졌다.

ABSTRACT

This paper describes procedures and the results of mineralogical, fabric and pore size analyses performed on four shales to understand time-dependent deformation behaviour of swelling shales better. Mineralogical compositions of these shales are established from the results of X-ray diffraction analysis and chemical analyses. The importance of the fabric in the understanding of swelling behaviour of shales is demonstrated using Scanning Electron Microscope (SEM). The change in pore size distribution during the process of swelling is investigated by measuring pore size distribution before and after free swell test.

The results of the present study imply that the swelling of shales studied is not attributed to minerals like pyrite, anhydrite or swelling clay minerals. The anisotropic swelling behaviour of shales studied may be explained by fabrics of these shales and the difference in them. The swelling of these shales is found to be accompanied by increase in the volume of pores.

INTRODUCTION

The time-dependent deformation behaviour of swelling rocks from
southern Ontario, Canada has been studied on fresh rockcores recovered
from boreholes drilled for different projects. The swelling rocks
studied in this research are all shales and they include shales from
Power Glen, Queenston, Georgian Bay and Blue Mountain Formations. The
list of these projects and rock formations studied is given in Table 1
and the locations of these projects are shown in Figure 1.

To understand time-dependent deformation behaviour better and study
swelling mechanism of shales, mineralogy and fabric of the above
mentioned shales have been studied in details. In addition, pore size
distribution of the Queenston shale was determined before and after
time-dependent deformation test to see if there is any change in pore
size distribution after the test. This paper describes procedures of
these analyses briefly and presents the results. Implications of the
findings from these analyses on time-dependent deformation behaviour
are then discussed.

ROCK FORMATIONS STUDIED

Among four shales studied, three shales are of the Upper Ordovician
Formations and they are the Blue Mountain, Georgian Bay and Queenston
shales. The other shale, the Power Glen shale, is of the Lower
Silurian Formation. The Blue Mountain Formation consists of grey to
brown fissile, thinly bedded soft shale. The dark brown shales,
ranging in thickness from 5 to 30 mm thick, are calcareous, while the
grey shales are generally noncalcareous. The Georgian Bay Formation
is mainly composed of soft, grey and brown, thinly bedded shale with
interbedded strong, grey limestone beds. The uppermost Ordovician
Queenston Formation is mainly a brick red shale with some green shale
partings or mottlings and scattered interbeds of limestone. The Power
Glen Formation consists of grey and greenish grey, laminated, fissile
sandy and calcareous shale with light grey fine-grained sandstone
interbeds throughout.

PROCEDURES OF MINERALOGICAL, FABRIC AND PORE SIZE ANALYSES

1. X-Ray Diffraction Analysis

All the X-ray diffraction studies have been carried out with a General Electric XRD-5 diffractometer using $CuK\alpha$ radiation generated at 50 kilovolts and 15 milliamperes. In general, all the X-ray traces are obtained at a range of 2000 C.P.S. with a scanning rate of 2° (2θ)/min. In order to identify the type of minerals present, X-ray diffraction analyses are made on untreated fines(random powder pattern) and oriented fines (either chemically pretreated or untreated). Generally, X-ray patterns are obtained on fines less than 74 μm fraction (for random pattern) and on oriented fines, prepared in distilled water and KCl solution and treated in various ways; wet, air-dried, ethylene glycol saturated and heated to 550 °C for 30 minutes.

The presence of vermiculite is established by potassium saturation which collapses the 1.42 nm peak to a 1.004 nm peak. Heat treatment to 550 °C for 30 minutes prior to X-raying is used to identify the chlorite and distinguish it from kaolinite. The disappearance of the 0.71 nm peak and a pronounced strengthening of the 1.42 nm peak indicate the presence of chlorite. Glycolation of air-dried specimen is used to identify the presence of any swelling minerals like smectite.

2. Chemical Analysis

The gasometric method using the Chittick apparatus developed by Dreimanis[1] is adopted to estimate the amount of calcite and dolomite. In this test, 6N HCl reacts with rock powder less than 74 μm and CO_2 gas is generated in this process. The volume of CO_2 gas generated is converted to the amount of calcite and dolomite using the standard calibration table.

The amount of quartz is quantitatively determined using the method of Gabriel Foscal-Mella[2]. In this method, a small amount (say 0.3 grams of less than No. 200 sieve) of potassium periodate(KIO_4) is added to and mixed thoroughly with rock powder less than 74 μm. From the X-ray traces on this sample, peak heights of quartz and KIO_4 at certain 2θ angles are compared to obtain the amount of quartz from a calibration curve with the known peak height ratio.

The amount of illite is quantitatively determined by measuring the potassium ion(K^+) present in the sample using a chemical method. In this method, potassium ion present in the sample is dissolved by washing the sample in several different acids. The percentage of K^+ is determined using the Atomic Absorption technique. Once the amount of K^+ is determined, the percentage of illite is obtained by assuming pure illite to have 6% of K^+.

The amount of vermiculite present is estimated from glycol retention analysis. This method consists of saturating rock powder with ethylene glycol, drying it in a dessicator and weighing it to constant weight. The amount of vermiculite is then estimated from the amounts of glycol retained in the untreated and the potassium saturated samples. The decrease in the glycol retention upon potassium saturation is attributed to the collapse of vermiculite to illite. If vermiculite is assumed to have a glycol retention of 150 mg/g, the amount of vermiculite present can then be roughly estimated from the reduction in glycol retention upon potassium saturation.

3. Fabric and Pore Size Analyses

Scanning Electron Microscope (SEM) photographs are taken for the fractured vertical or horizontal surfaces of a test specimen to study the effect of fabric on swelling behaviour qualitatively. The apparatus is the ISL DS-130 Dual Stage Scanning Electron Microscope in Surface Science Laboratory at The University of Western Ontario. The test specimen is prepared from the fresh fracture surface of air-dried samples. the specimen is cleaned with an air jet and then coated with gold-palladium. Then the specimen is ready for SEM work.

In order to study the effect of pore size distribution on swelling behaviour, pore size distribution is established using a porosimeter, Micromeritics Pore Sizer 9310. This apparatus is designed to measure small changes in volume of mercury, in which the porous material under study is immersed, when mercury is subjected to varying external pressures. Assuming pores to be spherical, the external pressure, p, is related to the equivalent pore diameter, d, as follows (Mitchell[3]) ;

$$d = - 4\tau \cos\theta \ / \ p$$

where τ is the surface tension of the intruding liquid and θ is the contact angle (for mercury, $\tau = 0.484$ N/m and $\theta = 141°$). As the pressure is increased, pores of smaller diameter are filled with the intruding mercury. The results are presented either in an incremental volume curve or a cumulative volume curve plotted against the equivalent pore diameter of the specimen.

RESULTS OF ANALYSES

1. Study of Mineralogy

In order to study the composition of shales, mineralogical and chemical analyses were performed on the Power Glen, Queenston, Georgian Bay and Blue Mountain shales. X-ray diffraction analysis was carried out to identify minerals present in these shales. For the quantitative determination of mineralogical composition, chemical analyses were carried out.

i) Qualitative Analysis

The results of X-ray diffraction analysis on random powder pattern of four swelling shales are shown in Figure 2. The X-ray powder pattern on the Queenston shale indicates that the Queenston shale contains abundant quartz and a minor amount of non-clay minerals such as calcite, dolomite and feldspar. Abundant clay minerals are also present as shown by strong peaks for both chlorite and illite. Similar observations can be made on the Power Glen shale and the Georgian Bay shale. However, the Blue Mountain shale contains much higher calcite than all the other shales studied, as shown by the strong 0.303 nm peak.

In Figure 3, X-ray traces of oriented fines are shown for natural sample and K^+ treated sample of Queenston shale. It may be observed from the results of natural sample that the strong peak at 1.004 nm reflects the presence of aboundant illite. The same strong peak at 1.004 nm is observed from all the other shales as reported in Lee [4]. From the traces obtained on the <74 μm fines heated to 550 °C, it can be noted that heat treatment destroyed the 0.71 nm peaks and enhanced the 1.42 nm peaks, indicating the presence of abundant chlorite. The presence of abundant chlorite is also observed from all the other shale samples except for the Blue Mountain shale. The presence of low angle clay minerals labelled "interlayered" is observed from traces for the oriented fines of the Queenston shale. The presence of interlayered clay minerals are also observed from the X-ray traces for the oriented fines of the Power Glen, Georgian Bay and Blue Mountain shales.

It is well known that the presence of vermiculite is reflected by the
increase in 1.004 nm peak at the expense of the 1.42 nm and 0.71 nm
peaks due to the collapse of vermiculite by potassium saturation. The
comparison of X-ray traces for air-dried states of untreated and K$^+$
saturated specimens indicates that vermiculite may not be present in
this shale, as reflected by the fact that potassium saturation does
not collapse 1.42 nm and 0.71 nm peaks and increase 1.004 nm peak.

From these mineralogical analyses, it is clear that the fines of all
four shales contain abundant quartz as non-clay mineral and abundant
illite and chlorite along with an assemblage of interlayered clays.

ii) Semi-Quantitative Analysis

The results of determination of mineralogical composition for four
shales are summarized in Table 2. These shale samples contain
abundant quartz ranging from 26 to 38%, while carbonates in these
shales are less abundant, amounting to 4 to 5%, except for the Blue
Mountain shale which contains 21% of carbonates. The amount of
feldspars reported in this Table is estimated from the ratio of the
peak heights of feldspar to calcite or dolomite. The amount of
feldspars determined in this way is in the range of 2 to 9%.

The approximate amount of illite present is obtained from the
percentage of K$^+$ after adjusting for the contribution of
K-feldspar. The amount of illite thus obtained ranges from 20 to 40%
for the four shales studied. In Table 3, the results of the glycol
retention test are summarized for the <74 μm fines of four shales.
The amount of glycol retained ranges from 18.5 to 20.9 mg/g for
untreated samples, while that for K$^+$ treated samples varies from
16.0 to 18.7 mg/g , giving the reduction in glycol of 1.6 to 4.2 mg/g.
The estimated vermiculite content from these reductions is in the
range of 1 to 3%. The rest of the <74 μm fines is considered to be
chlorite and interlayered clays, ranging from 8 to 29%.

It may be seen that only a trace of swelling clay mineral, in the form of vermiculite, is present. It is considered that these traces of vermiculite are insignificant to the swelling behaviour.

2. Study of Fabric

For the study of fabric, specimens of four shales were examined under the Scanning Electron Microscope (SEM). Photomicrographs of these four shales are shown in Plates 1, 2, 3, and 4.

In Plate 1, microscopic views of horizontal and vertical surfaces of the Georgian Bay shale are shown under two different magnifications; one at about 360 times and the other at about 4,000 times. The vertical surface of the Georgian Bay shale shows an assemblage of thin and wavy layers, which are edges of clay mineral platelets. These vertical surface photos reveal that the Georgian Bay shale has a strongly-layered fabric. This distinctive layering feature is however not well developed on the horizontal surface. The Blue Mountain shale shows a fabric with rather coarse, but well-developed layering on the vertical face, as shown in Plate 2. Very distinctive edges of clay mineral platelets may be noticed on the vertical surface. This strongly-layered fabric is also well demonstrated on the horizontal surface where all the domains(fabric units) are oriented to be face-on.

It may be observed from Plate 3 that the vertical surface of the Power Glen shale shows an assemblage of thin and slightly wavy layers, suggesting that the Power Glen shale has a strong clay fabric with preferred horizontal layering. This strong clay fabric is also well demonstrated on the horizontal surface where almost all the domains are oriented face-on. In the vertical surface of the Queenston shale (Plate 4), the layering is not well developed and there are a fair amount of domains and particles normal to horizontal layers. From the blow-up of vertical surface, it may be easily noticed that clay particles and domains are not well oriented to be edge-on. Similar observations may also be made from the horizontal surface. There is a weak trace of flaky layers, but domains and clay particles are generally oriented in a random manner.

Based on the above observations, it may be concluded that, among four shales studied, the Blue Mountain shale and the Power Glen shale show strongly-layered clay fabric, while the Queenston shale displays more random structure. The Georgian Bay shale shows a fabric in between these two.

3. Study of Pore Size Distribution

To evaluate the effect of the swelling on the structure of the shale, pore size distribution was determined on one sample after free swell test and a 'virgin' specimen (referred to as intact sample) of the Queenston shale. The results of pore size distribution analysis are shown in Figures 4 and 5.

In Figure 4, the cumulative pore volume of one 'intact' sample (IS) and one sample swelled for 520 days (FSS) is plotted against the equivalent pore diameter. The sample FSS shows more intrusion of mercury, indicating that this sample has more pore volume than the intact sample IS, as expected. In Figure 5, the incremental pore volume of samples IS and FSS is plotted against the equivalent pore diameter. Both samples have dominant pores in the ultra-micro range (less than 0.02 μm). The sample that swelled (FSS) shows an increase in volume of marco pores in the range of 100 to 300 μm and ultra-micro pores, implying that the swelling of shale is accompanied by an increase in the volume of macro pores and ultra-micro pores.

IMPLICATIONS OF THE FINDINGS ON SWELLING BEHAVIOUR OF SHALES

It is well known that some minerals like pyrite, anhydrite and swelling clay minerals (e.g. vermiculite) are responsible for the time-dependent deformation behaviour of argillaceous materials, e.g. clay, shale, marl and anhydrite (details of these swelling mechanisms may be found Lee [4] and elsewhere). However, the results of mineralogical analysis show that time-dependent deformation of the shales studied may not be attributed to the presence of minerals like pyrite, anhydrite and swelling clay minerals (e.g. vermiculite).

The results of fabric analysis indicate that the shales studied
generally show a clay fabric with some horizontal layering, though the
degree of layering varies from one shale to another. The effect of
fabric on macro structure of shales is well demonstrated in Plate 5.
This Plate shows photos of broken rock fragments of Power Glen shale
and Queenston shale. It may be seen that the Power Glen shale was
broken to predominantly platy shape upon drying and wetting,
reflecting the effect of strongly-layered fabric. On the other hand,
the Queenston shale was deteriorated to sub-round rock fragments,
showing the influence of the more random fabric.

The effect of fabric on swelling of shales is manifested in the fact
that the vertical swelling under free swell test condition is normally
higher than the horizontal swelling, though the stress relieved in the
vertical direction is generally smaller than that in the horizontal
direction (Lee[5]). The degree of horizontal layering seems to
govern the anisotropy of swelling in the vertical and horizontal
directions. This may be demonstrated by using the ratio of swelling
potential in the vertical direction to that in the horizontal
direction. The Blue Mountain shale, having a very strong clay fabric,
gives the average ratio of 6.3 (from 2 samples), indicating that the
vertical swelling is about 6.3 times higher than the horizontal
swelling. On the other hand, the vertical swelling potential of the
Queenston shale, showing random fabric, is only about 1.6 times higher
than the horizontal swelling potential (from 9 tests) as reported in
Lee [5]. The Georgian Bay shale, exhibiting a fabric intermediate
between the above two extremes, shows an average of 4.3 (from 14
tests), revealing the effect of the intermediate fabric.

CONCLUSIONS

To understand time-dependent deformation behaviour of shales studied,
a comprehensive program for mineralogical, fabric and pore size
analyses was carried out for the Blue Mountain shale, the Georgian Bay
shale, the Queenston shale and the Power Glen shale. From the results
of these analyses, the following conclusions may be drawn :

(a) The shales studied generally contain abundant clay minerals, but
    only traces of swelling clay minerals in the form of vermiculite.
    There is no evidence of pyrite or anhydrite in these shales,
    implying that the swelling of shales studied can not be attributed
    to minerals like pyrite, anhydrite or swelling clay minerals.

(b) Both the Blue Mountain shale and the Power Glen shale show a
    strong clay fabric with perferred horizontal layering, while the
    Queenston shale displays a rather randomly oriented fabric. The
    Georgian Bay shale shows a fabric in between these two. The
    results of fabric study and observations on swelling potential
    measured suggest that the anisotropic swelling behaviour of shales
    may be explained by different degrees of horizontal layering.

(c) The results of pore size analysis show that an increase in the
    volume of macro pores (100 to 300μ m) and ultra micro pores
    (less than 0.02 μ m) occurs during the swelling of shales.

REFERENCES

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   dolomite using Chittick Apparatus, <u>J. Sediment Petrol.</u>,
   Vol. 32, 1962, PP 520 ~ 529

2. Foscal-Mella, G., <u>Maitrise es Sciences Appliquees</u>,
   Ecole Polytechnique, 1976

3. Mitchell, J. K., <u>Fundamentals of Soil Behaviour</u>, John Wiley & Sons
   Inc., 1976

4. Lee, Y.N., <u>Stress-Strain-Time Relationship of Queenston Shale</u>, Ph.
   Thesis, Univ. of Western Ontario, London, Canada, 1988

5. Lee, Y.N., Time-dependent deformation behaviour of Queenston shale,
   <u>J. Korean Society of Geotechnical Engineers</u>, Vol. 5, No.2,
   1989, PP 57 ~ 72

TABLE 1 LIST OF PROJECTS AND ROCK FORMATIONS STUDIED

Project	Location	Year of Study	Excavation Type	Time-Dependent Deformation Study		
				Rock Formation	Depth of Sampling (m)	Orientation of Specimen
Scotia Plaza	Toronto	1984	Open	Georgian Bay Shale	<30	H
Skydome Stadium	Toronto	1985	Open	Georgian Bay Shale	<30	V,H [1]
Lakeview Deephole	Mississauga	1985	-	Georgian Bay & Blue Mountain Shale	0 - 270	V
John St. Storm Outfall Tunnel	Toronto	1986	Tunnel	Georgian Bay Shale	<30	H
SAGNGS No. 3	Niagara Falls	1984	Tunnel	Grimsby, Power Glen & Queenston shale	>40	H
SABNGS No. 3	"	1985	"	Queenston Shale	≈95	V, HM, HN [2]
SABNGS No. 3	"	1986	",	"	≈105	V, HM, HN
SABNGS No. 3	"	1987	"	"	≈110	V, HM, HN
Hamilton	Hamilton	1987	"	"	<30	V, H

NOTE: [1] H stands for horizontal, V for vertical

[2] HM and HN stand for the major and minor horizontal stress directions respectively for oriented specimens.

TABLE 2   SUMMARY OF MINERALOGICAL COMPOSITION OF THE <74 μm FINES

Minerals % Sample No.	Calcite	Dolomite	Quartz	Feldspar	Illite	Vermiculite	Chlorite & Inter-layered Clays
SAB3/FS/PG2 (Power Glen Shale)	2	3	38	3	27	2	25
SAB3/FS/Q1 (Queenston Shale)	2	2	26	2	40	1	27
LUDH/FS7 (Georgian Bay Shale)	2	2	37	9	20	1	29
LUDH/FS12 (Blue Mt. Shale)	18	3	28	8	32	3	8

TABLE 3   RESULTS OF GLYCOL RETENTION TESTS ON <74 µm FINES

Sample No.	Glycol Retained (mg/g)		Reduction in Glycol Retained Upon K^+ Saturation (mg/g)	% Vermiculite
	Untreated <74 µm	Homo K^+ <74 µm		
SAB3/FS/PG2	18.5	16.0	2.5	1.7
SAB3/FS/Q1	19.5	17.6	1.9	1.3
LUDH/FS7	20.3	18.7	1.6	1.1
LUDH/FS12	20.9	16.7	4.2	2.8

Note: In the estimation of vermiculite content, the amount of glycol retained in
       vermiculite was assumed to be 150 mg/g.
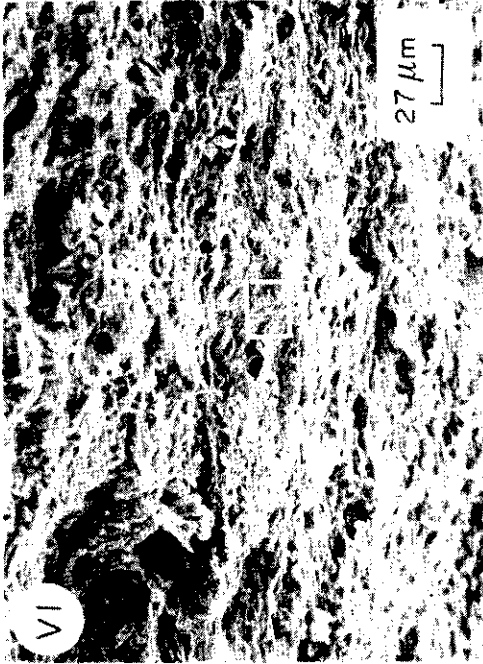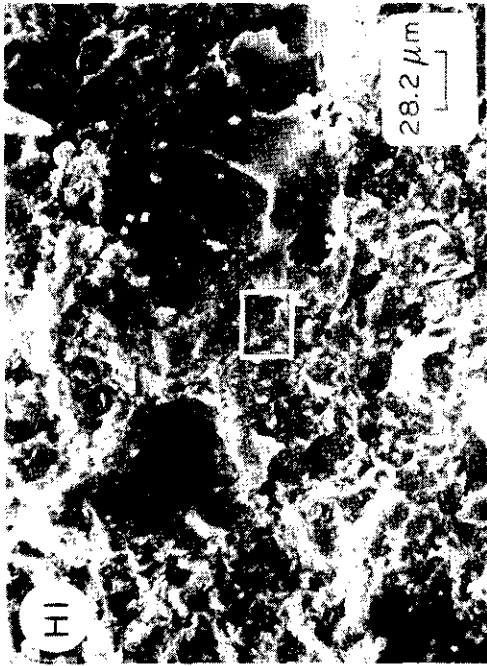
VERTICAL SURFACE

HORIZONTAL SURFACE

PLATE 1   SCANNING ELECTRON PHOTOMICROGRAPHS OF THE
GEORGIAN BAY SHALE, AIR-DRIED

V1

25.6 μm

V2

2.6 μm

H1

25.6 μm

H2

2.0 μm
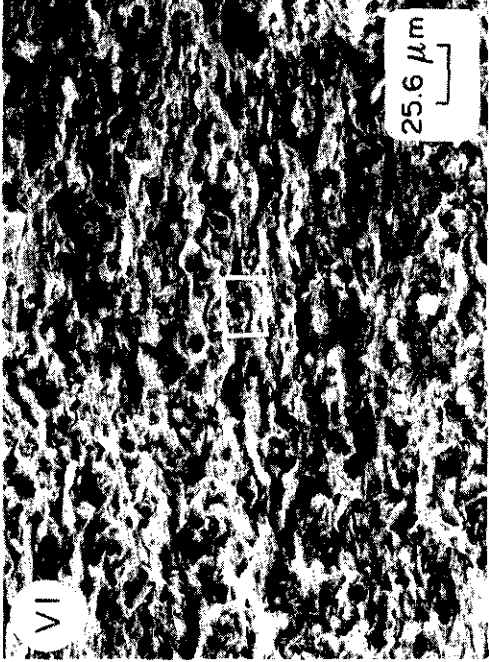
VERTICAL SURFACE

HORIZONTAL SURFACE

PLATE 2   SCANNING ELECTRON PHOTOMICROGRAPHS OF THE
BLUE MOUNTAIN SHALE, AIR-DRIED

V1 — 28 μm

V2 — 2.8 μm

VERTICAL SURFACE

H1 — 25.6 μm

H2 — 2.6 μm
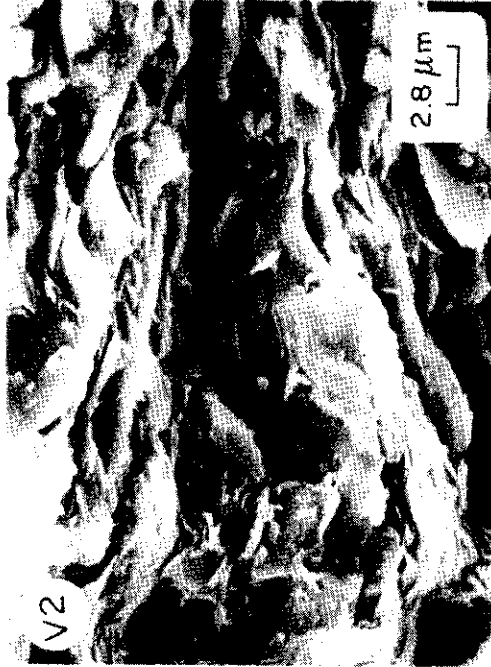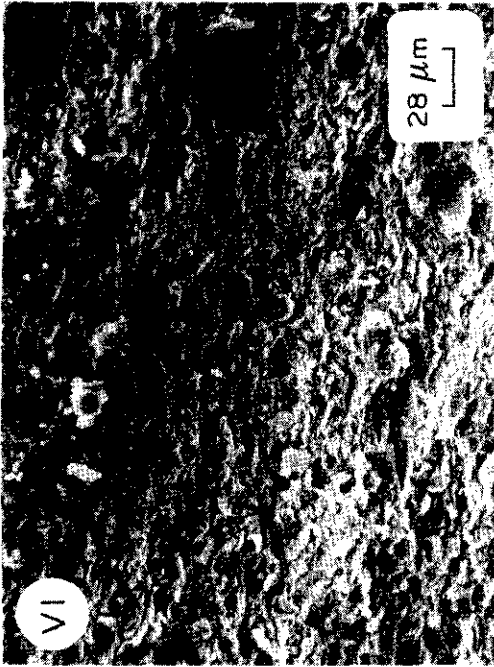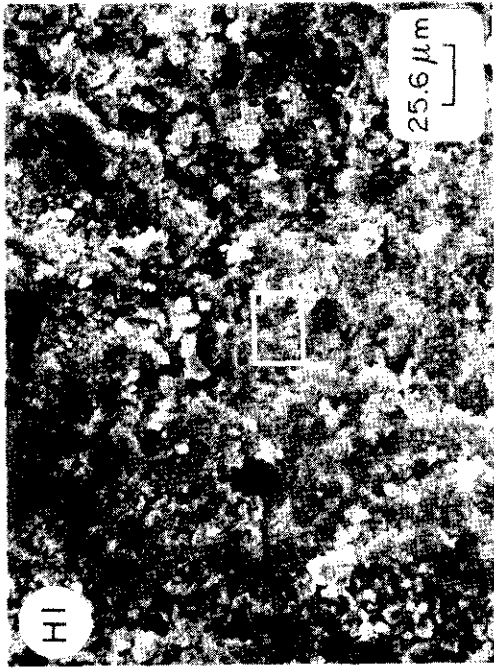
HORIZONTAL SURFACE

PLATE 3   SCANNING ELECTRON PHOTOMICROGRAPHS OF THE
POWER GLEN SHALE, AIR-DRIED
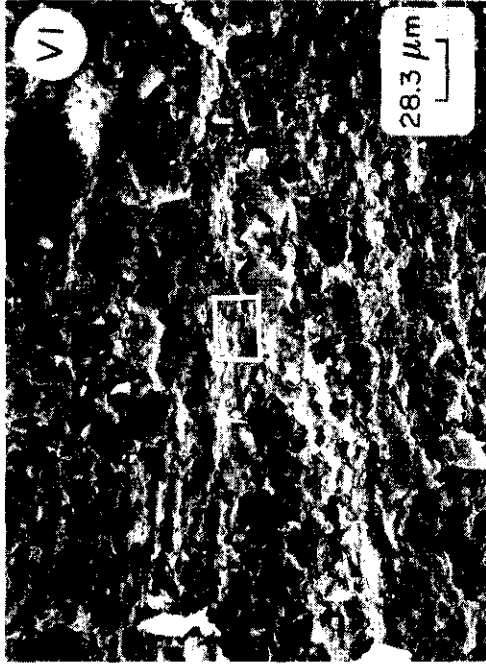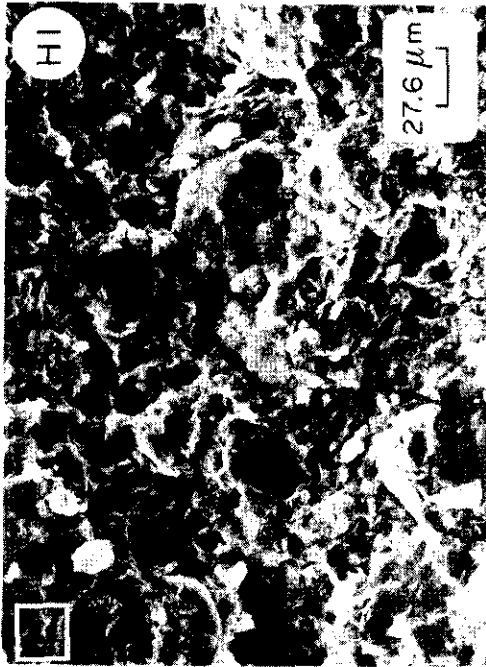
VERTICAL SURFACE

HORIZONTAL SURFACE

PLATE 4   SCANNING ELECTRON PHOTOMICROGRAPHS OF THE
QUEENSTON SHALE, AIR-DRIED

(a)
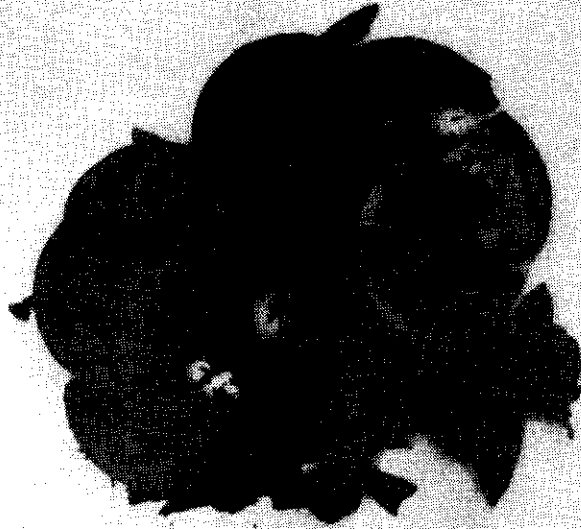
(b)

PLATE 5   DETERIORATED SHALE SAMPLES UPON DRYING
            AND REWETTING; (a) POWER GLEN SHALE
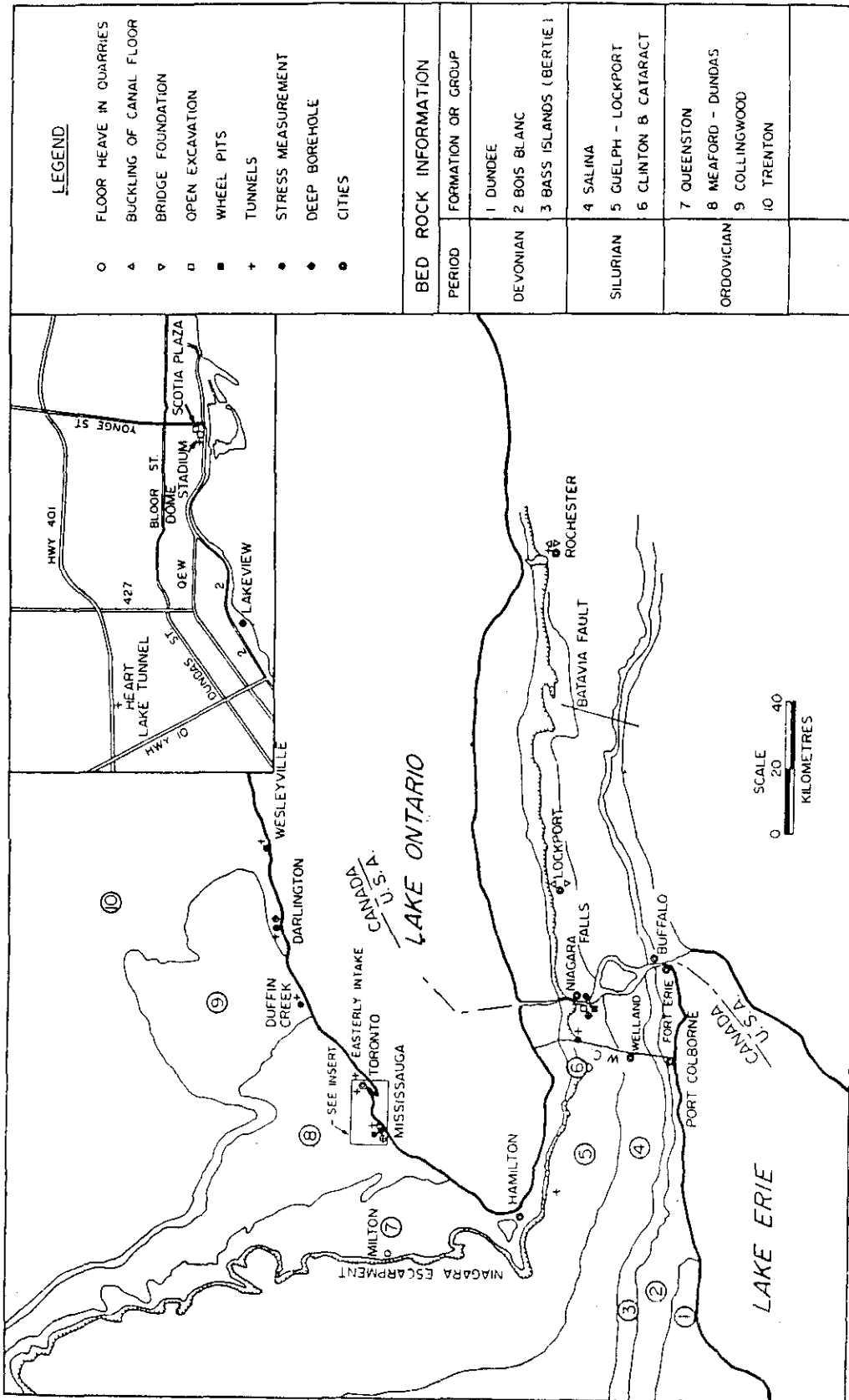   AND (b) QUEENSTON SHALE

FIGURE 1 GEOLOGICAL CONDITION, LOCATION OF MAJOR STRUCTURES AND STRESS MEASUREMENTS (From Lo, 1986)
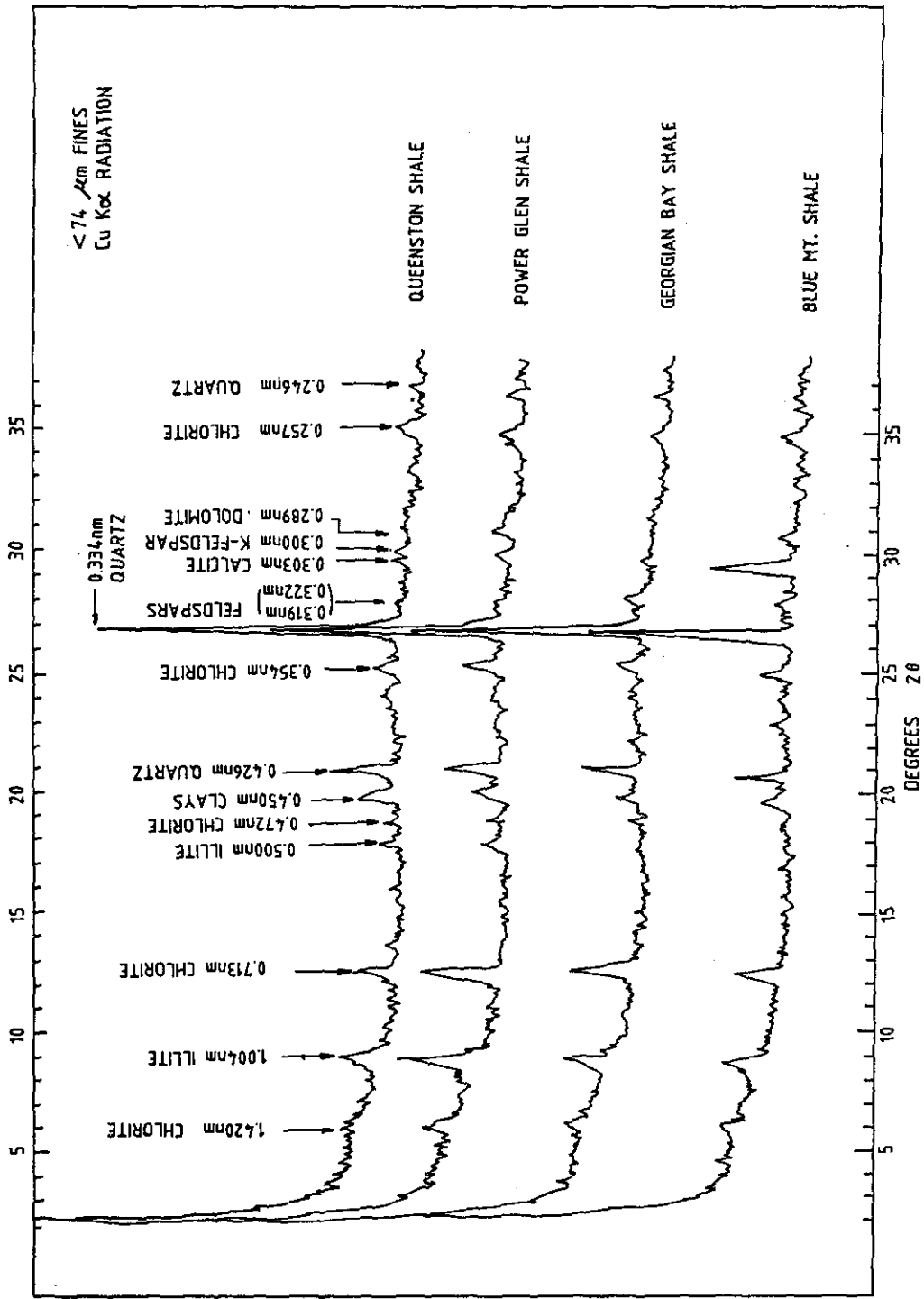
FIGURE 2   X – RAY POWDER PATTERN OF   FOUR SWELLING SHALES FROM SOUTHERN ONTARIO, CANADA
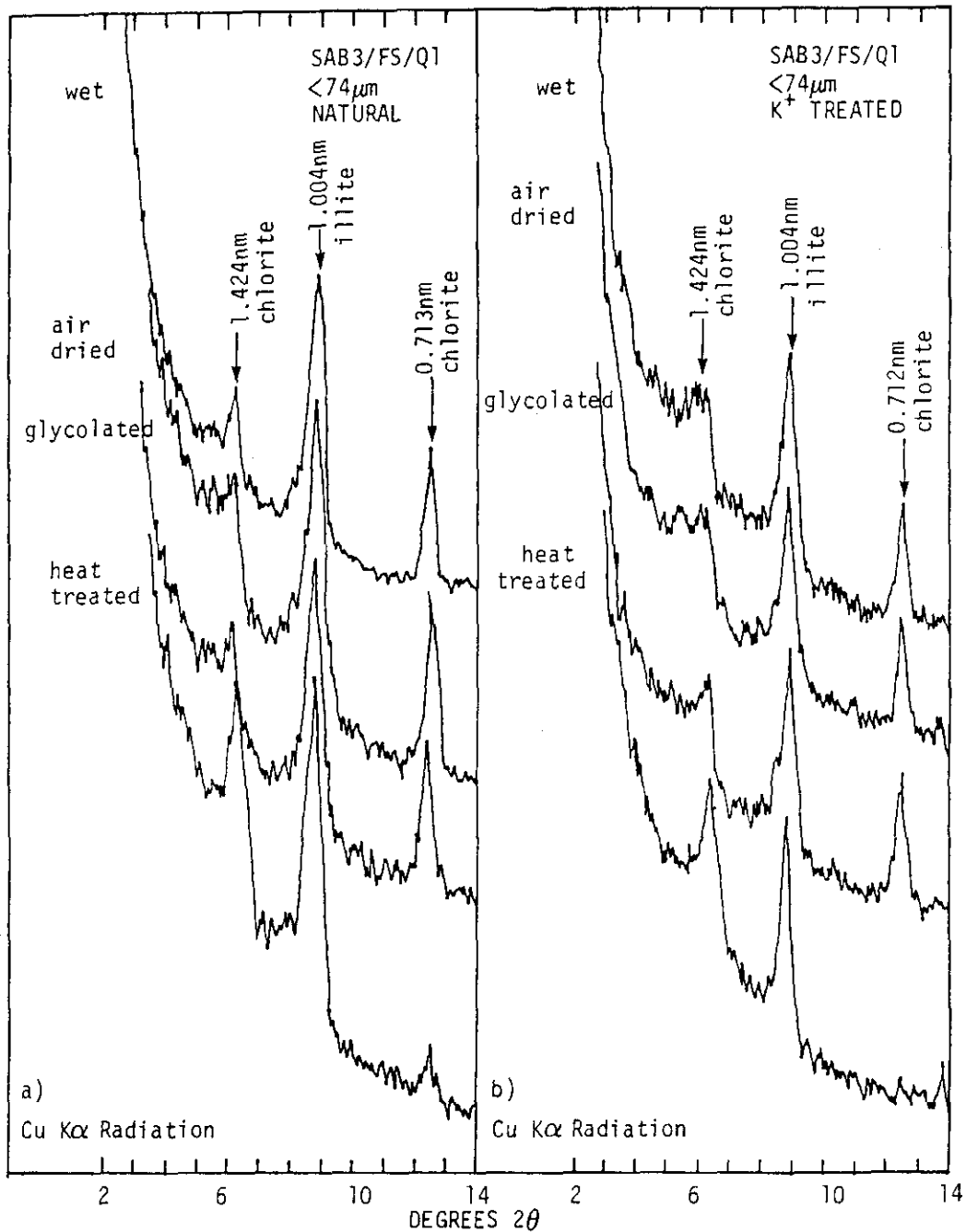
FIGURE 3 X-RAY DIFFRACTION TRACES OF ORIENTED, <74 μm FINES, QUEENSTON SHALE, NIAGARA FALLS; (a) NATURAL SAMPLE AND (b) K⁺ TREATED SAMPLE

TEST NO	SAMPLE CONDITION	CALCITE CONTENT(%)	W.C.[1] (%)	V.S.P.[2]
SAB3(83)/IS	INTACT	4.2	2.70	0
SAB3(86)/FSS	FREE-SWELLED	5.7	3.45	0.4

NOTES: (1) W.C. stands for water content measured at the given sample condition

(2) V.S.P. stands for vertical swelling potential in %/log cycle of time

SAB3(83)/IS

SAB3(86)/FSS

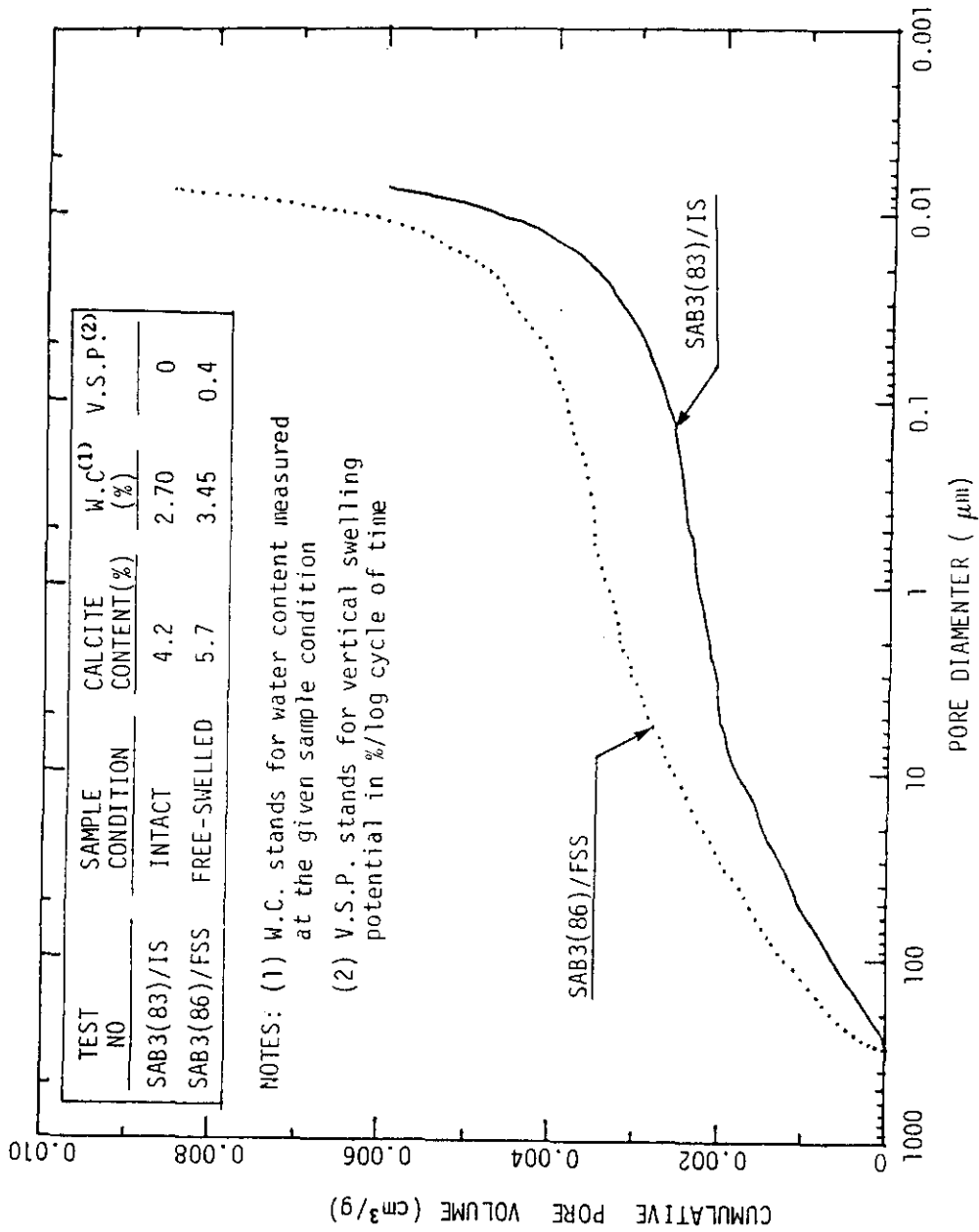CUMULATIVE PORE VOLUME (cm³/g)

PORE DIAMENTER (μm)

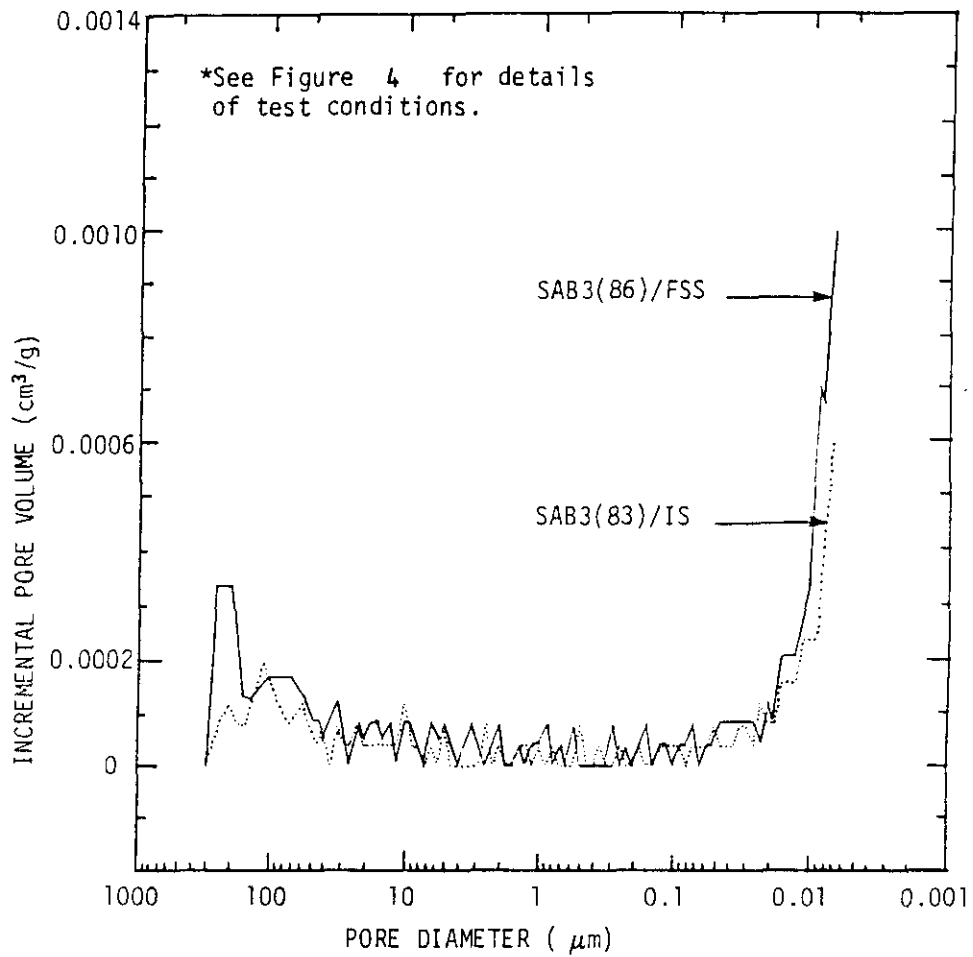FIGURE 4 CUMULATIVE PORE SIZE DISTRIBUTION OF TWO QUEENSTON SHALE SAMPLES

FIGURE 5   INCREMENTAL PORE SIZE DISTRIBUTION FOR
TWO QUEENSTON SHALE SAMPLES