Soil and Groundwater Contamination

-Determining the transport properties of low permeability geological materials in the laboratory-

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Abstract

The accurate characterization of transport properties of low permeability environments has important practical implications, such as in the fields of soil and water contamination and geological disposal of radioactive nuclear waste. Although principal mechanisms of mass and/or fluid transport in geologic media include advection, dispersion, chemical reaction, chain decay and/or biochemical retardation, etc, the most fundamental phenomena are advection and dispersion. When fluid flow is very flow, such as in the case of flow in low permeability environment, the dispersion becomes all most equivalent to diffusion. The advection phenomenon basically related to the hydraulic or flow properties, and the dispersion phenomenon basically related to the diffusive transport properties can be evaluated from the permeability and diffusion tests, respectively. Although characteristics of geologic media are generally fractured and heterogeneous, and tests can also be performed in situ, this talk will concentrate only on laboratory tests on continuous and homogeneous geologic materials in the laboratory, due to the limit of time.

Although several types of laboratory permeability and diffusion tests are available and have been widely used in geotechnical and/or geo-environmental fields, traditional test methods based on simple solutions corresponding to simplified boundary conditions have limitations in testing low permeability geo-materials, either due to the need of very long testing time and/or low accuracy.

This talk will be divided into two parts. In part I, the speaker will review recent advances in theoretical analyses of laboratory permeability tests, present potential strategies for effectively decreasing the test time, introduce a new and versatile laboratory system which can implement any of 6 test methods and show a series of experimental results which demonstrate the accuracy and efficiency of the new laboratory system. In part II, the speaker will review the concept and theory of conventional through-diffusion test, indicate potential problems, present 2 rigorous solutions to the through-diffusion test, theoretically evaluate the applicability and limitations of conventional through-diffusion test, and show an example of improved technique. Considerations and approaches presented in this talk may offer basic ideas to avoid misinterpreting the experimental data.

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APEC Seminar
GSJ, AIST, 071129

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Determining the transport properties of low-permeability geological materials in the laboratory

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Outline

The accurate characterization of transport properties of low-K Environments has important practical implications:

*Soil and Groundwater Contamination
*Geological disposal of radioactive nuclear wastes

Principal mechanisms of mass/fluid transport in geologic media

*Advection	Hydraulic/flow properties	Permeability tests	PART I
*Dispersion	Diffusive transport properties	Diffusion tests	PART II

*Chemical reaction, *Chain decay, *Biochemical retardation, etc.

Characteristics of geologic media	Test methods
Fractured Heterogeneous	In-situ
Continuous Homogeneous geologic materials	In laboratory

Part I: Laboratory permeability tests

1) Review advancements in theoretical analyses of 3 techniques
   (Const.-Head, Const.-Flow Rate & Transient-Pulse K Tests)
2) Present potential strategies for effectively decreasing the time
   (Required to estimate the low-K)
3) Introduce a new and versatile laboratory system
   (Can implement any of the 3 test methods)
4) Show a series of experimental results
   Demonstrate the accuracy and efficiency
5) Re-classification & further developments of lab. K tests
   6 kinds of test methods and rigorous solutions to them
6) Further improvements in laboratory system
   Automation of lab. system operation & data analyses

* Lab. Permeability Tests and Theories *

4 Laboratory Techniques

- Constant-Head(CH)
- Falling-Head(FH*)

Measuring induced flow rates

(Estimate the K of A Saturated Specimen)

- Const. Flow-Rate(FP)
- Transient-Pulse(TP)

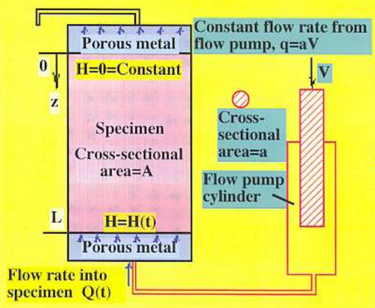
Measuring time-dependent variations of differential hydraulic head

*A special case of a TP permeability test

• Cont. F. R. Technique as an Example

* Lab. Permeability Tests and Theories *

Constant Flow-Rate



Schematic diagram

Governing Equ.

$$\frac{\partial^2 H}{\partial z^2} - \frac{S_S}{K} \cdot \frac{\partial H}{\partial t} = 0$$

Initial condition

$$H = 0 \quad at \quad 0 \le z \le L$$

Boundary condition

$$H = 0 \quad at \quad z = 0$$

$$\frac{\partial H}{\partial z} = \frac{1}{KA}\left(q - C_e \frac{\partial H}{\partial t} \right) \quad at \quad z = L$$

$Q(t)$

➡ Rigorous Analytical Solution

The initial condition: hydraulic head within the specimen before a test is zero;
Boundary conditions: at the outflow end the hydraulic head is kept to be zero;
at the inflow end, the actual flow rate into specimen Q(t) equals the constant flow rate from flow pump q minus the volume absorbed within the permeating system per unit time interval due to the increase in hydraulic head within it. Ce: equipment compliance.

* Lab. Permeability Tests and Theories *

Rigorous analytical solution

$$H = \frac{qL}{AK}\left[\frac{z}{L} - 2 \sum_{n=0}^{\infty} \frac{exp\left(-\frac{K}{S_S}\beta_n^2 t \right) sin\left(\beta_n z\right)}{L\delta\beta_n cos\left(\beta_n L\right)\left[L\left(\beta_n^2 + \frac{1}{\delta^2}\right) + \frac{1}{\delta} \right]} \right]$$

Darcy's Law

$$\delta = \frac{C_e}{AS_s} \qquad tan\left(\beta L\right) = \frac{1}{\beta\delta}$$

Morin and Olsen (1987)

$$H = \frac{qL}{AK}\left[1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{exp\left(-\frac{K}{S_S}(2n+1)^2 \pi^2 t / 4L^2 \right)}{(2n+1)^2} \right]$$

➡ Hydraulic Gradient Distribution

Allow z=L & disregard the Ce, the solution reduces to the expression as developed by Dr. Morin & Dr. Olsen. A large times, simplifies to the steady state expression as defined by Darcy's Law.

* Lab. Permeability Tests and Theories *

Rigorous analytical solution

$$H=\frac{qL}{AK}\left\{\frac{z}{L}-2\sum_{n=0}^{\infty}\frac{\exp\left(-\frac{K}{S_S}\beta_n^2 t\right)\sin\left(\beta_n z\right)}{L\delta\beta_n\cos\left(\beta_n L\right)\left[L\left(\beta_n^2+\frac{1}{\delta^2}\right)+\frac{1}{\delta}\right]}\right\}$$

$$\delta=\frac{C_e}{AS_s} \qquad \tan\left(\beta L\right)=\frac{1}{\beta\delta}$$

Hydraulic gradient distribution

$$i(z,t)=\frac{qL}{AK}\left\{\frac{1}{L}-2\sum_{n=0}^{\infty}\frac{\exp\left(-\frac{K}{S_S}\beta_n^2 t\right)\cos\left(\beta_n z\right)}{L\delta\cdot\cos\left(\beta_n L\right)\left[L\left(\beta_n^2+\frac{1}{\delta^2}\right)+\frac{1}{\delta}\right]}\right\}$$

➡ All Solutions

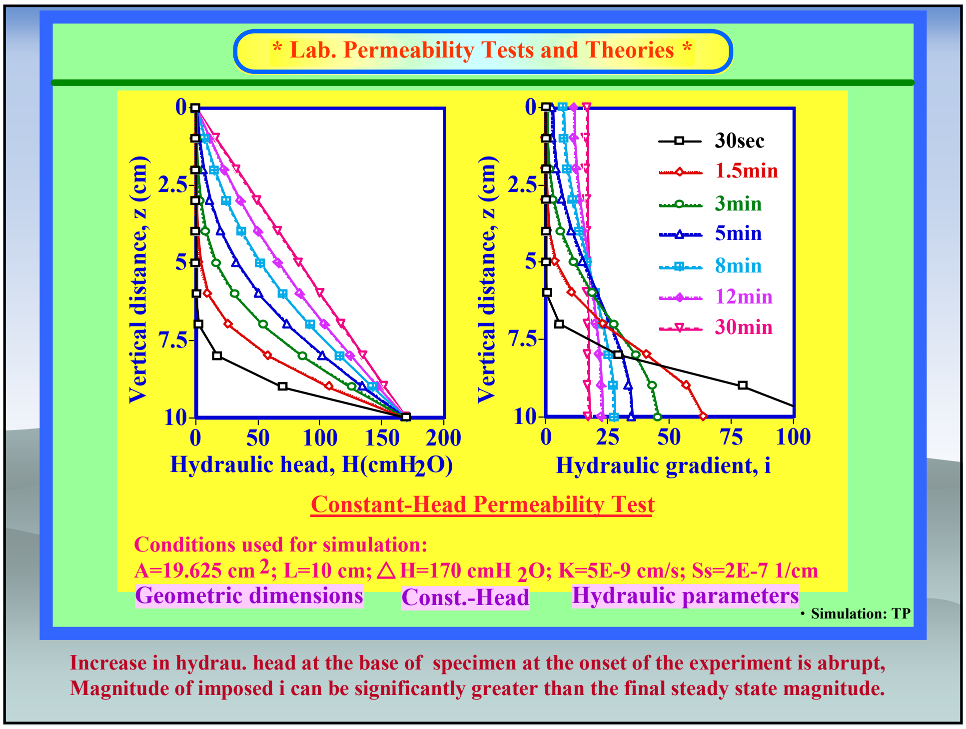
i **can be obtained by differentiating the analytical solution with respect to the variable z**
Maximum value of ***i*** **during a test can be controlled by q of the pump.**

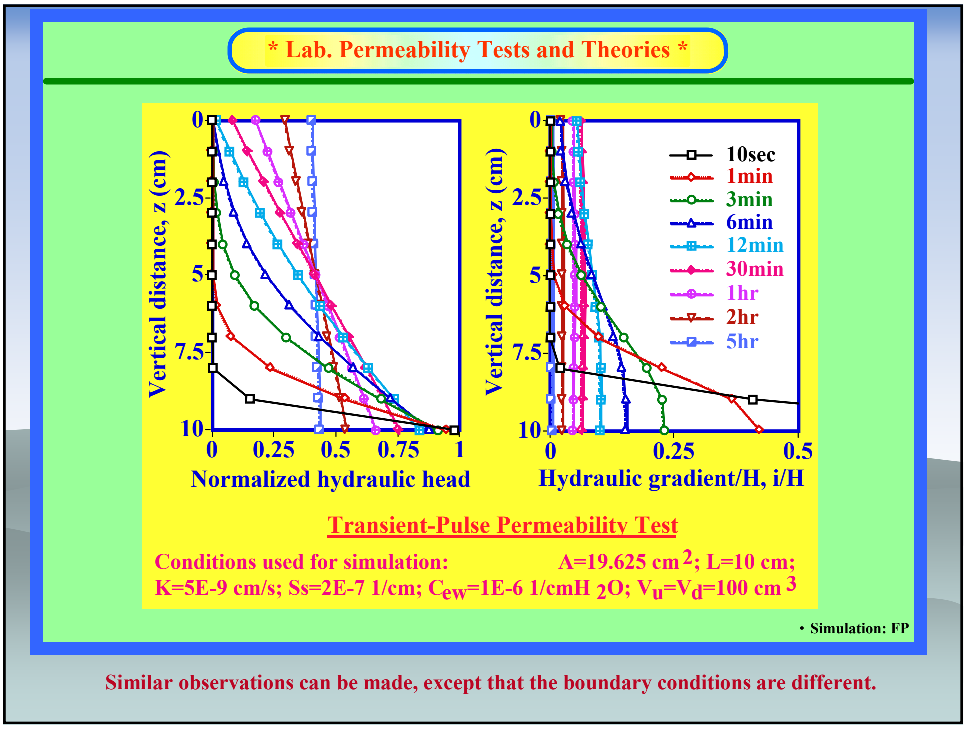
* Lab. Permeability Tests and Theories *

Method	Steady-S Exp.	Hydraulic head	Hydraulic gradient
Constant head	$K=-\frac{q}{A\cdot i}=-\frac{ql}{A\cdot\Delta H}$	$H(z,t)=\Delta H\left\{\frac{z}{L}+\frac{2}{\pi}\sum_{n=1}^{\infty}\frac{\cos(n\pi)}{n}\cdot\sin\frac{n\pi z}{L}\cdot\exp\left(-\frac{K}{S_s}\cdot\frac{n^2\pi^2}{L^2}t\right)\right\}$	$i(z,t)=\frac{\Delta H}{L}\left\{1+2\sum_{n=1}^{\infty}\cos(n\pi)\cdot\cos\left(\frac{n\pi z}{L}\right)\cdot\exp\left(-\frac{K}{S_s}\cdot\frac{n^2\pi^2}{L^2}t\right)\right\}$
		Zhang et al. 1998, Geotechnical Testing J., 21(1), 52-57.	
Constant flow-rate	$K=-\frac{q}{A\cdot i}=-\frac{ql}{A\cdot\Delta H}$ Olsen, 1966, Water Res. Res., 2(6), 287-295.	$H(z,t)=\frac{qL}{AK}\left\{\frac{z}{L}-2\sum_{n=0}^{\infty}\frac{\exp\left(-\frac{K}{S_s}\beta_n^2 t\right)\sin\left(\beta_n z\right)}{L\delta\beta_n\cos\left(\beta_n L\right)\left[L\left(\beta_n^2+\frac{1}{\delta^2}\right)+\frac{1}{\delta}\right]}\right\}$ $\delta=\frac{C_e}{A\cdot S_s}$ $\tan\left(\beta L\right)=\frac{1}{\beta\delta}$	$i(z,t)=\frac{qL}{AK}\left\{\frac{1}{L}-2\sum_{n=0}^{\infty}\frac{\exp\left(-\frac{K}{S_s}\beta_n^2 t\right)\cos\left(\beta_n z\right)}{L\delta\cdot\cos\left(\beta_n L\right)\left[L\left(\beta_n^2+\frac{1}{\delta^2}\right)+\frac{1}{\delta}\right]}\right\}$
		Esaki et al. 1996, Geotech. Testing J., 19(3), 241-246. Zhang et al. 1998, Geotechnical Testing J., 21(1), 52-	
Transient pulse	$h_u-h_f=H\cdot\frac{V_d}{V_u+V_d}\cdot e^{-\alpha t}$ $K=\frac{\theta\rho\lambda lV_uV_d}{A(V_u+V_d)}$ Brace et al., 1968, JGR, 73(6), 2225-2236.	$\frac{H(z,t)}{H}=\frac{1}{1+\beta+\gamma}+2\sum_{m=0}^{\infty}\times\frac{\exp(-\alpha\phi_m^2)\left[\cos\phi_m\xi-(\gamma\phi_m/\beta)\sin\phi_m\xi\right]}{\left(1+\beta+\gamma-\gamma\phi_m^2/\beta\right)\cos\phi_m-\phi_m\left(1+\gamma+2\gamma/\beta\right)\sin\phi_m}$ $\xi=\frac{z}{L},\ \alpha=\frac{Kt}{L^2S_s},\ \beta=\frac{S_sAL}{S_u},\ \gamma=\frac{S_d}{S_u}$ $\tan\phi=\frac{(\gamma+1)\phi}{\gamma\phi^2/\beta-\beta}$	$\frac{i(z,t)}{H}=-2\sum_{m=0}^{\infty}\frac{\phi_m}{L}\times\frac{\exp\left(-\frac{Kt}{L^2S_s}\phi_m^2\right)\left[\sin\left(\phi_m\frac{z}{L}\right)-\left(\frac{\gamma\phi_m}{\beta}\right)\cos\left(\phi_m\frac{z}{L}\right)\right]}{\left(1+\beta+\gamma-\frac{\gamma\phi_m^2}{\beta}\right)\cos\phi_m-\phi_m\left(1+\gamma+\frac{2\gamma}{\beta}\right)\sin\phi_m}$
		Hsieh, et al. 1981, Int. J. Rock Mech. Min. Sci., 18(3), 245-252.	Zhang et al. 2000, Geotechnical Testing J., 23(1), 83-99.

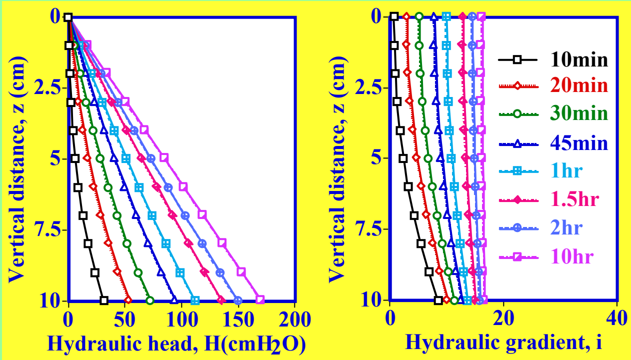
➡ Simulation: constant head

Using the rigorous analytical solutions together with properly set conditions, particular response characteristics of each technique can be simulated and evaluated.


* Lab. Permeability Tests and Theories *
Vertical distance, z (cm)
Hydraulic head, H(cmH2O)
Hydraulic gradient, i
30sec
1.5min
3min
5min
8min
12min
30min
Constant-Head Permeability Test
Conditions used for simulation:
A=19.625 cm 2; L=10 cm; △H=170 cmH 2O; K=5E-9 cm/s; Ss=2E-7 1/cm
Geometric dimensions
Const.-Head
Hydraulic parameters
• Simulation: TP
Increase in hydrau. head at the base of specimen at the onset of the experiment is abrupt,
Magnitude of imposed i can be significantly greater than the final steady state magnitude.


* Lab. Permeability Tests and Theories *
Vertical distance, z (cm)
Normalized hydraulic head
Hydraulic gradient/H, i/H
10sec
1min
3min
6min
12min
30min
1hr
2hr
5hr
Transient-Pulse Permeability Test
Conditions used for simulation:
A=19.625 cm 2; L=10 cm;
K=5E-9 cm/s; Ss=2E-7 1/cm; Cew=1E-6 1/cmH 2O; Vu=Vd=100 cm 3
• Simulation: FP
Similar observations can be made, except that the boundary conditions are different.

* Lab. Permeability Tests and Theories *



Constant Flow-Rate Permeability Test

Conditions used for simulation: A=19.625 cm^2; L=10 cm; K=5E-9 cm/s; Ss=2E-7 1/cm; q=0.1 μ L/min; C_e=2E-5 cm^2

Control the maxim. *i*

• Simulation: Summary

At the onset of the experiment, the increase in hydr. head at the base of the specimen is gradual
i are established slowly across the entire specimen until a steady-state bound is reached
that is also the maximum *i* controlled by the flow rate of the pump.

* Lab. Permeability Tests and Theories *

Systematically changing the values of selected parameters

- Effects on individual lab. tests

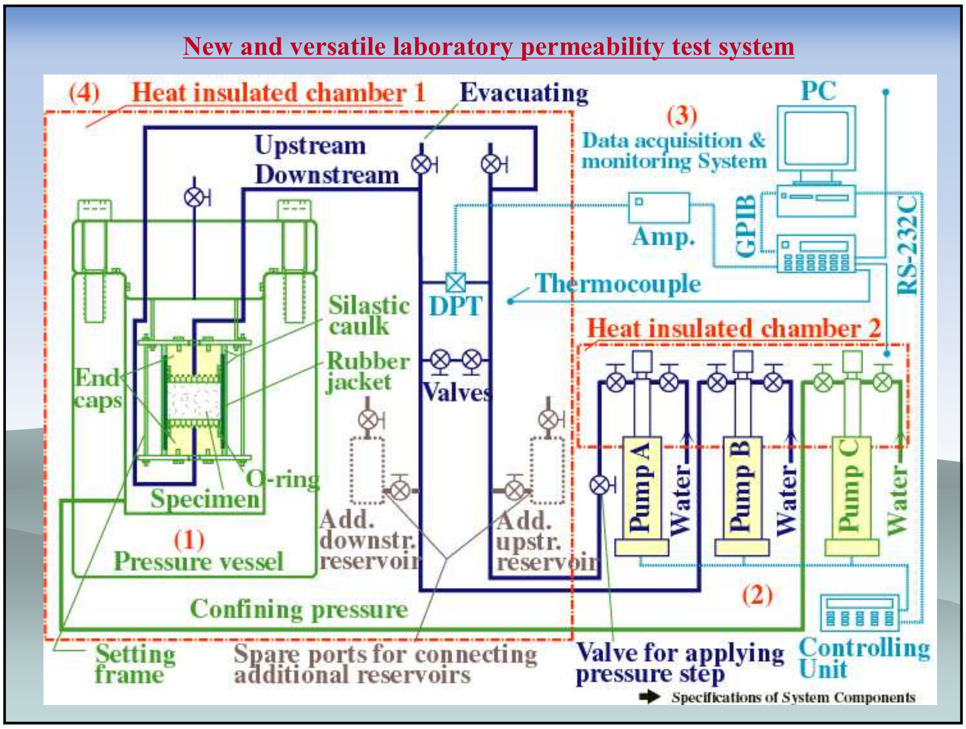
Major observations:

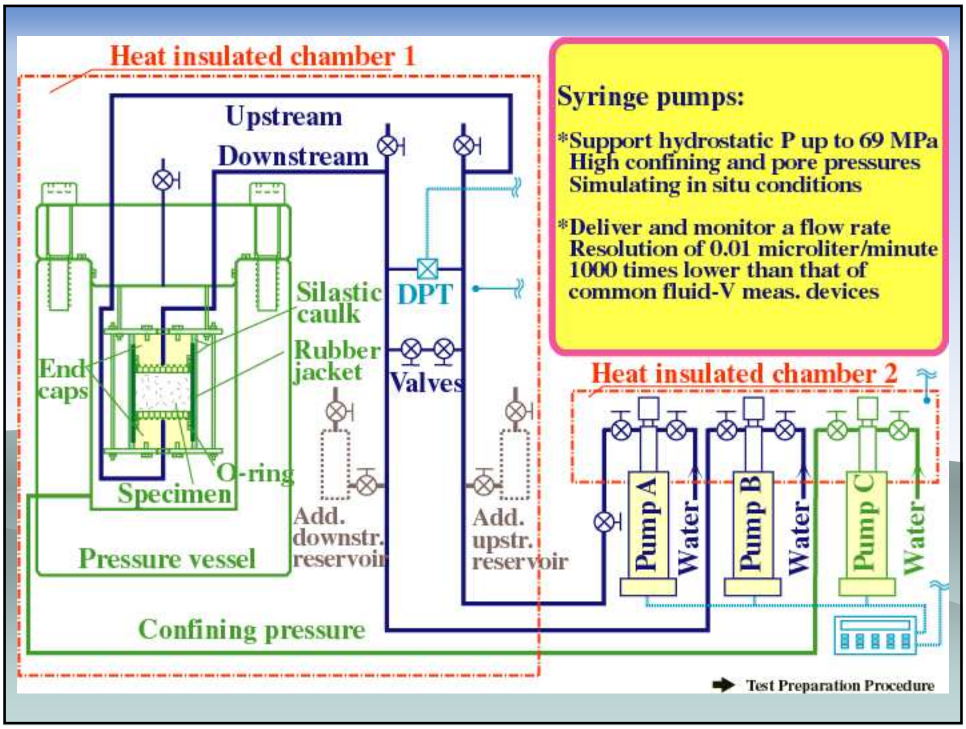
- To shorten the T required to measure low K
  To use a shorter specimen having a larger A

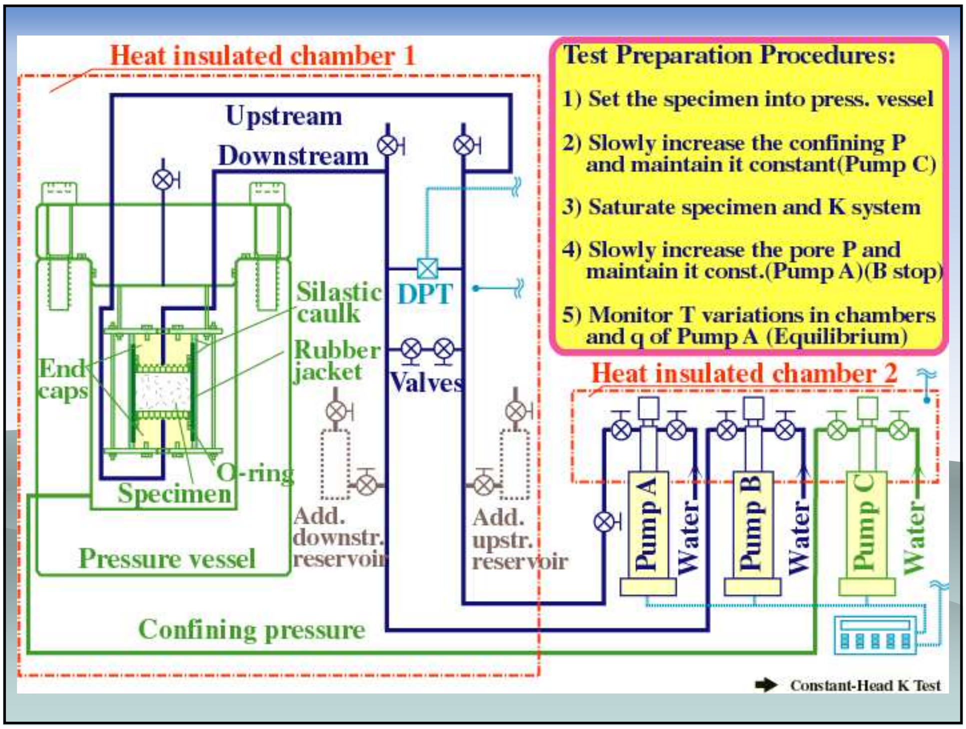
New criteria for determining the optimum specimen size
*The miximum grain size
*The miximum length of micro-crack

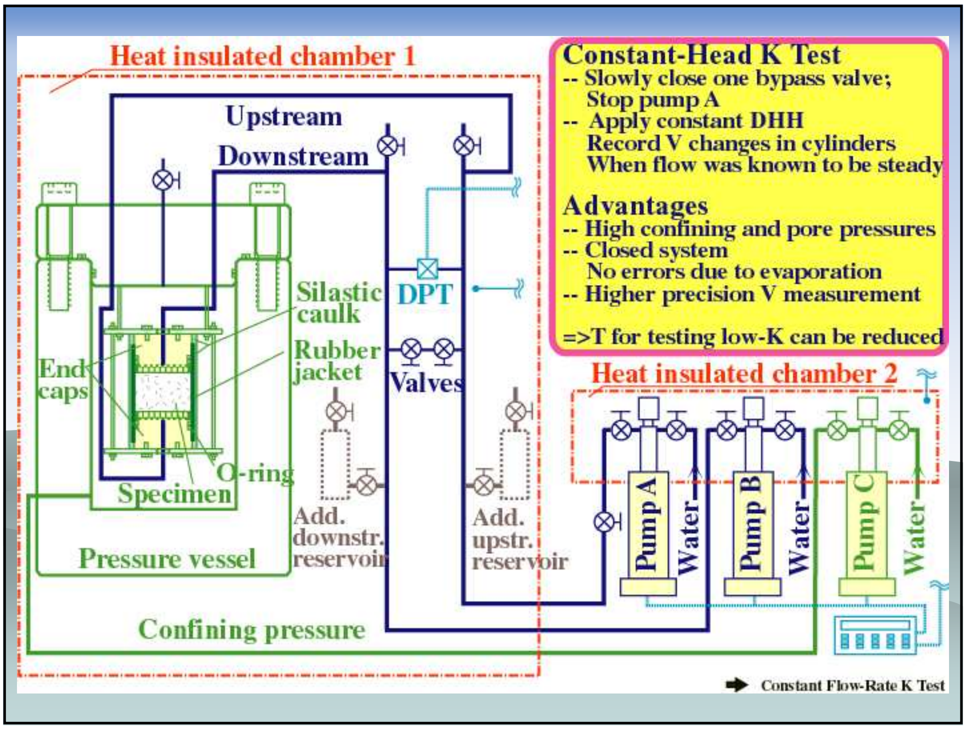
D, L of specimen > 5-10 times the physical parameters ?!

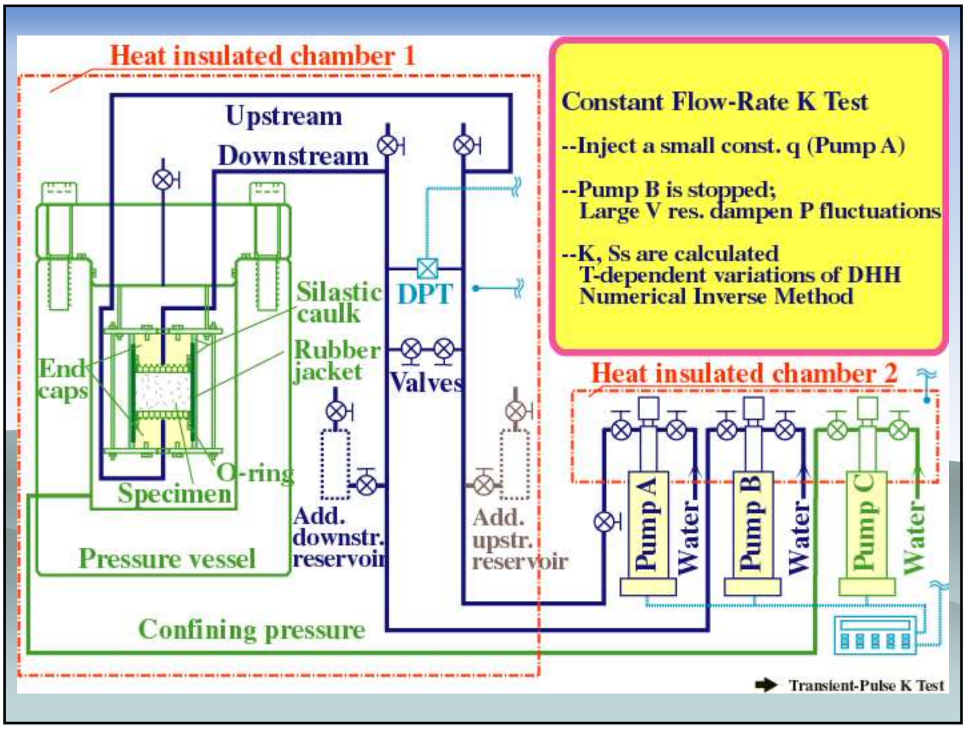
Omit Numerical Inverse Method: • New & Versatile Lab. K Test System

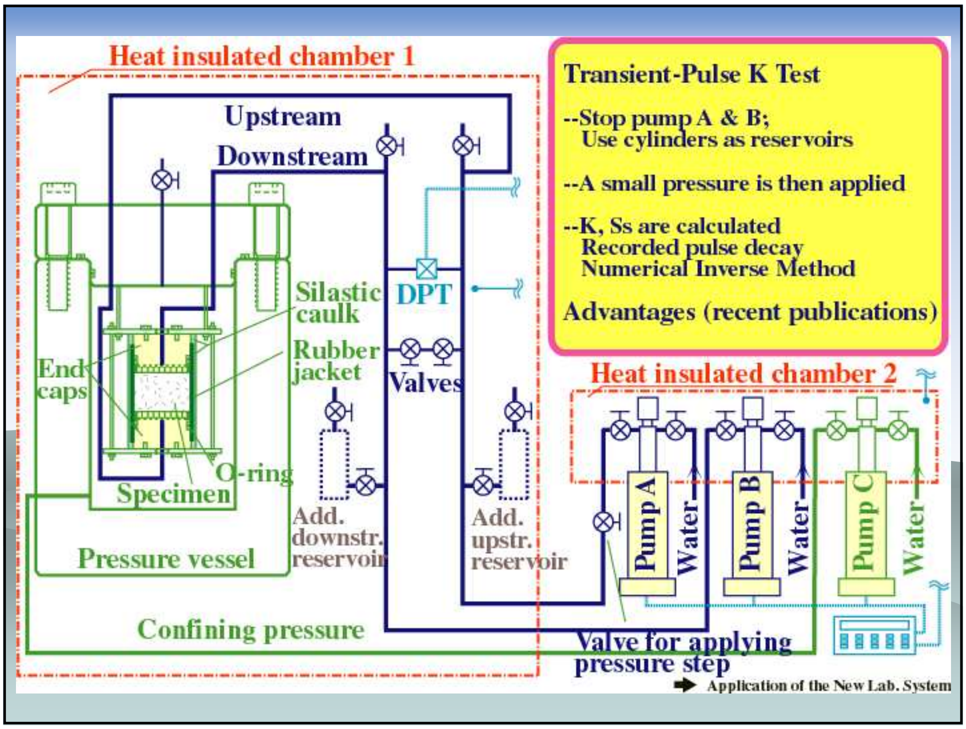

New and versatile laboratory permeability test system
(4) Heat insulated chamber 1
Evacuating
Upstream
Downstream
PC
(3)
Data acquisition & monitoring System
Amp.
GPIB
RS-232C
Thermocouple
DPT
Silastic caulk
Rubber jacket
Valves
End caps
O-ring
Specimen
(1)
Pressure vessel
Add. downstr. reservoir
Add. upstr. reservoir
Heat insulated chamber 2
Pump A
Water
Pump B
Water
Pump C
Water
(2)
Confining pressure
Setting frame
Spare ports for connecting additional reservoirs
Valve for applying pressure step
Controlling Unit
Specifications of System Components

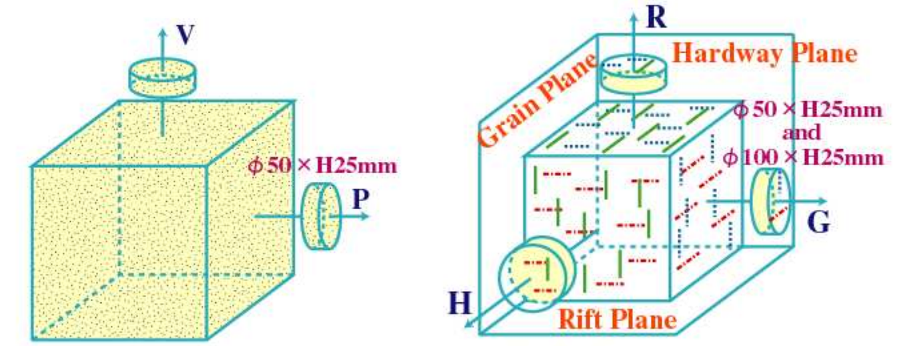

Heat insulated chamber 1
Upstream
Downstream
DPT
Silastic caulk
Rubber jacket
Valves
End caps
O-ring
Specimen
Pressure vessel
Add. downstr. reservoir
Add. upstr. reservoir
Confining pressure
Syringe pumps:
*Support hydrostatic P up to 69 MPa
High confining and pore pressures
Simulating in situ conditions
*Deliver and monitor a flow rate
Resolution of 0.01 microliter/minute
1000 times lower than that of
common fluid-V meas. devices
Heat insulated chamber 2
Pump A
Water
Pump B
Water
Pump C
Water
Test Preparation Procedure


Heat insulated chamber 1
Upstream
Downstream
Silastic caulk
DPT
Rubber jacket
Valves
End caps
O-ring
Specimen
Add. downstr. reservoir
Add. upstr. reservoir
Pressure vessel
Confining pressure
Test Preparation Procedures:
1) Set the specimen into press. vessel
2) Slowly increase the confining P and maintain it constant(Pump C)
3) Saturate specimen and K system
4) Slowly increase the pore P and maintain it const.(Pump A)(B stop)
5) Monitor T variations in chambers and q of Pump A (Equilibrium)
Heat insulated chamber 2
Pump A
Water
Pump B
Water
Pump C
Water
Constant-Head K Test


Heat insulated chamber 1
Upstream
Downstream
Silastic caulk
DPT
Rubber jacket
Valves
End caps
O-ring
Specimen
Add. downstr. reservoir
Add. upstr. reservoir
Pressure vessel
Confining pressure
Constant-Head K Test
-- Slowly close one bypass valve; Stop pump A
-- Apply constant DHH
Record V changes in cylinders
When flow was known to be steady
Advantages
-- High confining and pore pressures
-- Closed system
No errors due to evaporation
-- Higher precision V measurement
=>T for testing low-K can be reduced
Heat insulated chamber 2
Pump A
Water
Pump B
Water
Pump C
Water
Constant Flow-Rate K Test


Heat insulated chamber 1
Upstream
Downstream
Silastic caulk
DPT
Rubber jacket
Valves
End caps
O-ring
Specimen
Pressure vessel
Add. downstr. reservoir
Add. upstr. reservoir
Confining pressure
Constant Flow-Rate K Test
--Inject a small const. q (Pump A)
--Pump B is stopped;
Large V res. dampen P fluctuations
--K, Ss are calculated
T-dependent variations of DHH
Numerical Inverse Method
Heat insulated chamber 2
Pump A
Water
Pump B
Water
Pump C
Water
Transient-Pulse K Test


Heat insulated chamber 1
Upstream
Downstream
Silastic caulk
DPT
Rubber jacket
Valves
End caps
O-ring
Specimen
Pressure vessel
Add. downstr. reservoir
Add. upstr. reservoir
Confining pressure
Transient-Pulse K Test
--Stop pump A & B;
Use cylinders as reservoirs
--A small pressure is then applied
--K, Ss are calculated
Recorded pulse decay
Numerical Inverse Method
Advantages (recent publications)
Heat insulated chamber 2
Pump A
Water
Pump B
Water
Pump C
Water
Valve for applying pressure step
Application of the New Lab. System



a) Shirahama sandstone
(Sedimentary rock)

b) Inada granite
(Igneous rock)

- To investigate the effects of heterogenity on rock K
  Parallel to and perpendicular to bedding for sandstone
  Perpendicular to Rift, Grain and Hardway Planes for granite
- Disc-shaped cylindrical specimens (see Figs) were used
  To reduce the time required for a test.

➡ Testing Strategies

* Application of the New Lab. System *

Testing Strategies

- To evaluate the capabilities of the new lab. system
- To cross-check the results obtained from the 3 methods
  Sandstone (SVS) & Granite (GHL)
  A series of confining & pore P conditions
- To investigate the effects of confining P & rock heterogenity
  A series of K tests was performed
  Specimens cored in different orientations for both rocks
  Confining P was increased stepwise up to 60 MPa
  A TP test was conducted at each step

• Conditions for Cross-check K Tests; Test No.

* Application of the New Lab. System *

Part of the Conditions

Table 4 Conditions for cross-check K tests.

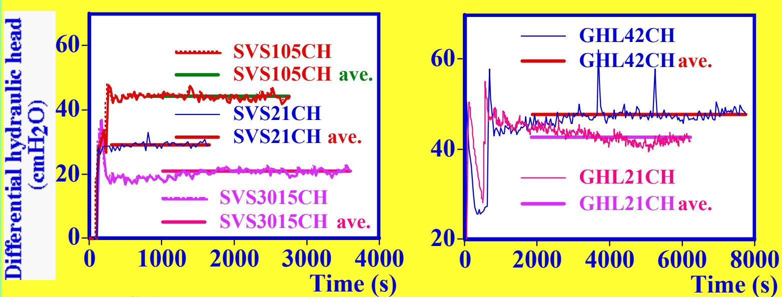
Shirahama sandstone				Inada granite			
Test No.	Conf. P (MPa)	Pore P (MPa)	Other condi.*	Test No.	Conf. P (MPa)	Pore P (MPa)	Other cond.*
SVS21CH SVS21FP SVS21TP	2	1	29.1 0.005 68.4	GHL1.5CH GHL1.5FP GHL1.5TP	1	0.5	29.4 0.002 34.9
•	•	•	•	•	•	•	•
SVS4020CH SVS4020TP	40	20	31.4 76.2	GHL302FP GHL302TP	30	2	0.0002 117.7

Rock type: S-sandstone; G-granite
Orientation: V, P-Perpend. to & parall. to bedding for sandstone
R,G,H-Perpend. to Rift, Grain & Hardway Planes for granite
Specimen size: S-D50 X H25mm; L-D100 X H25mm
Pressure conditions: Confining and pore P in MPa
Test method: CH-Constant Head; FP-Constant Flow Rate(Flow Pump); TP-Transient Pulse

*Other conditions: Hydraulic head in cmH_2O for CH; q in cm^3/min for FP; Pulse pressure in cmH_2O for TP

• Examples of CH Results

* Application of the New Lab. System*



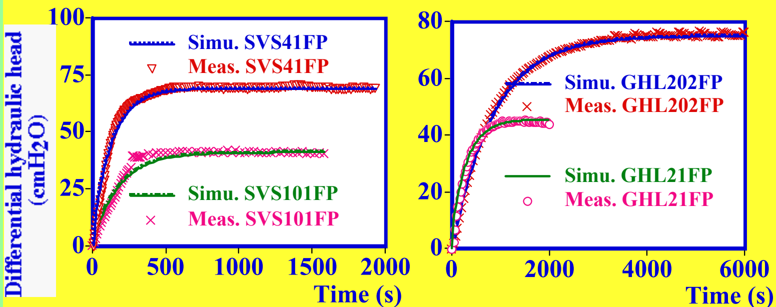
a) Shirahama sandstone b) Inada granite

Constant-Head Permeability Test

*Each curve illustrates the T-dependent variations in DHH across the specimen
*After a certain period of T, the DHH approaches a quasi-steady state
*Changes in inflow & outflow volumes are measured over a certain T span
*The values of K are calculated from the inflow & outflow rates and their ave.

• Examples of FP Results

* Application of the New Lab. System*



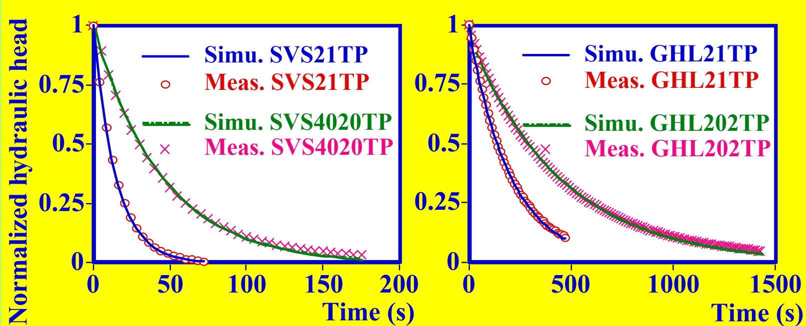
a) Shirahama sandstone b) Inada granite

Constant Flow-Rate Permeability Test

*The curves illustrate the T-dependent rise in DHH
Symbols & lines depict the meas. & simu. results
*K & Ss are calculated from whole-T and half-T measurements
K from only the steady-state measurements are also calculated
*Simulated curves are in close agreement with experimental data
The accuracy of the rigorous theoretical analysis for this test

• Examples of TP Results

* Application of the New Lab. System*



a) Shirahama sandstone b) Inada granite

Transient-Pulse Permeability Test

*The figures depict the decay of the normalized DHH
Symbols & lines are the meas. & simu. results
*K & Ss are calculated from the meas. data
By means of the numerical inverse method
*Simu. curves are in close agreement with exp. data
The accuracy of the rigorous theoretical analysis

• Results Table

* Application of the New Lab. System*

Table 5 Results obtained from the cross-check tests. (T= 15 ℃)

	Test No.	Constant-head				Constant-flow rate(FP)				Trans.-pulse	
		K_i	K_o	K_{ave}	Time	K_h	K_w	K_s	Time	K	Time
Sand-stone		Omit				Omit				Omit	
Granite	GHL1.5	3.06	4.29	3.68	3 240	4.57	4.51	4.53	2 970	5.91	792
	GHL21	4.14	3.10	3.62	5 775	4.28	4.77	4.85	1 980	6.61	470
	GHL42	3.51	3.28	3.40	5 788	3.88	4.10	4.11	2 790	7.12	940
	...	...	...	...	...	...	...	...	...	...	...

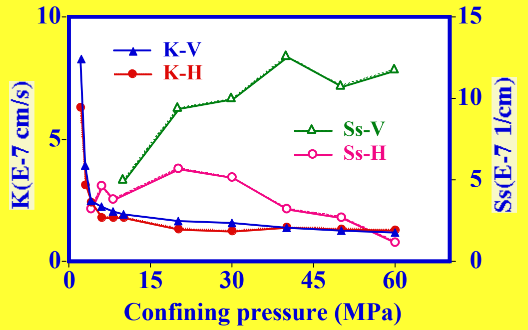
K : ($\times 10^{-9}$ cm/s); Time: s; K_i & K_o : K obtained from inflow & outflow;
h, w & s: Half-time, whole-time and steady-state

*K values obtained from the three tests are on the same order
- Measuring substantial changes in hydr. P at early T in TP test?
- Very small plumbing leaks?
- Slight temperature variations?

*K from half-T & whole-T estimation during a FP test are the same
In close agreement with the steady-state estimation
=> Numerical inverse method based on rigorous theoretical analysis permits to reduce the T required for testing low-K specimens

• Effects of Pc on K, Ss of Shirahama sandstone

* Application of the New Lab. System*

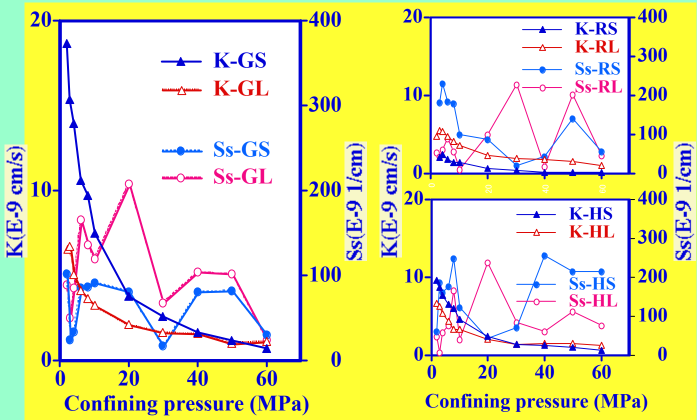


Effects of confining P & heterogeneity on K & Ss of Shirahama sandstone.

• Effects of Pc on K and Ss of Inada granite

The tests were performed under Pp be kept at 1 Mpa and the Pc increased stepwise up to 60 Mpa.
K ranged from about 8 to 1 E-7 in cm per second, and Ss ranged from about 13 to 1 E-7 in per cm, respectively.
The reduction in K is due to the closure of microcracks with increment in confining pressure.
The variation in Ss may be due to a cyclical generation and closure of secondary microcracks formed perpend. to the contacting surface between grains during hydrostatic crushing.
The K in the directions perpendicular and parallel to bedding are almost the same.
This implies that Shirahama sandstone is relatively isotropic in a hydraulic sense.

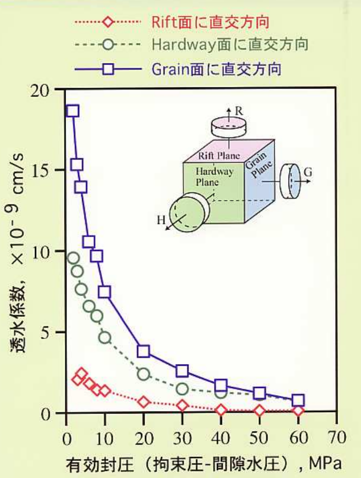
*** Application of the New Lab. System***



Effects of confining P and heterogeneity on K and Ss of Inada granite.

• Conclusion

Ranges are about two orders lower than those of Shirahama sandstone.



稲田花崗岩の計測例

Permeabilities in the three directions perpendicular to Grain, Rift and Hardway planes are different, especially in the direction perpendicular to Rift Plane.
This implies that Inada granite is an isotropic geologic material.

* Summary and Conclusions*

*To reduce the time required to measure low-K
- Use a disk-shaped specimen
  - Max. grain size & max. length of microcracks
  - Incorporated to developing a new standard

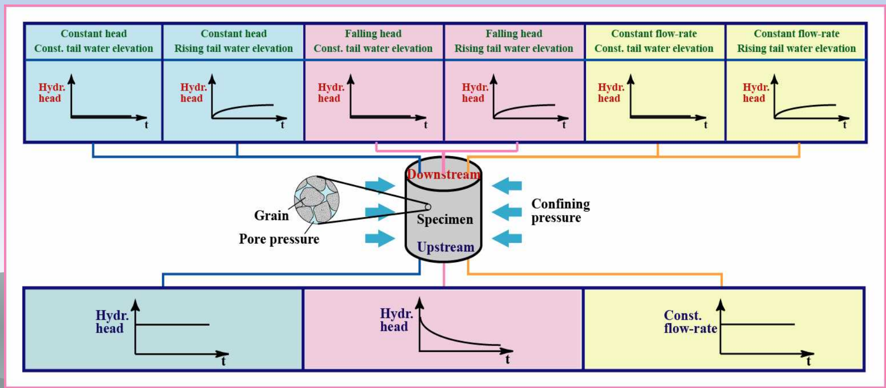
*Numerical Inverse Method
- Based on rigorous theoretical analyses
- Permits the test duration to be significantly shorted
- Without sacrificing accuracy in estamating both K & Ss

*The new lab. system
- An effective & versatile equipment for testing low-K
- K derived from the 3 complementary methods are in agreement

*K & Ss of a rock are functions of confining pressure
- K decreases monotonically with increasing stress
- Ss varies circularly

Re-classification & further developments of lab. K tests

Re-classification of lab. K tests



Constant-Head

Falling-Head Transient Pulse

Constant Flow-Rate

Re-classification & further developments of lab. K tests

Rigorous solutions to the other 3 tests are now available

Upstream Boundary:
Constant-head

Downstream Boundary:
Rising tail water elevation

Upstream Boundary:
Falling-head

Downstream Boundary:
Constant tail water elevation

Zhang, M., et. al., 2004, Simultaneous Determination of the Hydraulic Conductivity and Specific Storage of A Test Specimen From Laboratory Permeability Tests, Eos Trans. AGU, 85(47), Fall Meet. Suppl., Abstract H21E-1065.

Upstream Boundary:
Constant flow-rate

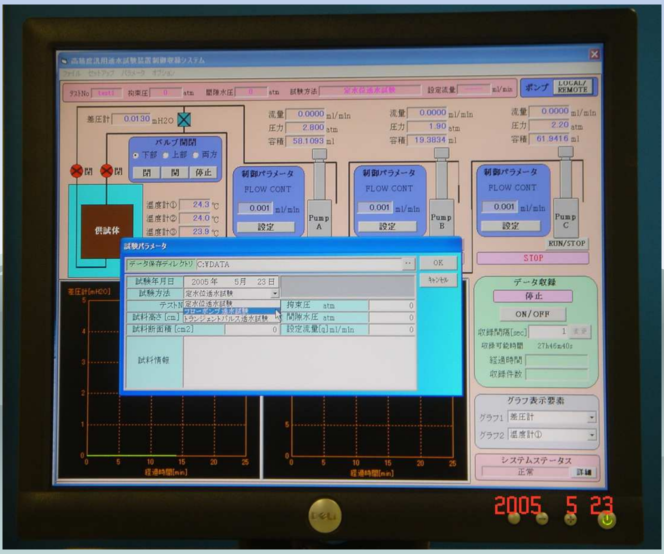
Downstream Boundary:
Rising tail water elevation

Song, I., et. al., 2004, One-Dimensional Fluid Diffusion Induced by Constant-Rate Flow Injection: Theoretical Analysis and Application to the Determination of Fluid Permeability and Specific Storage of a Cored Rock Sample, Journal of Geophysical Research - Solid Earth, 109 (B5): doi:10.1029/2003JB002395.

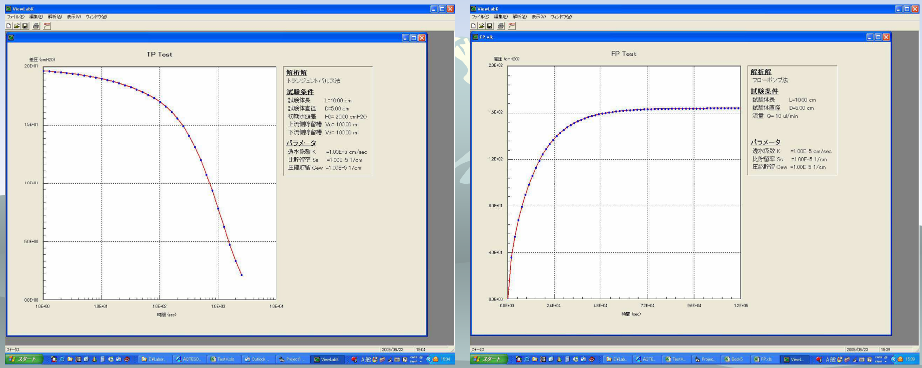
Further improvements in laboratory system

Picture of Automated Versatile Laboratory K System

Further improvements in laboratory system
Automated system operation



Further improvements in laboratory system
Automated data analyses



Part II: Laboratory diffusion tests

Lab. diffusion test is a well-established & widely adopted approach
Characterizing the diffusive transport properties of geo-materials
Several types of diffusion tests currently available
Through-diffusion has been extensively used for testing rock samples

Contents of today's talk

1) Review concept and theory of conventional through-diffusion test
Indicate potential problems

2) Present 2 rigorous solutions to through-diffusion test

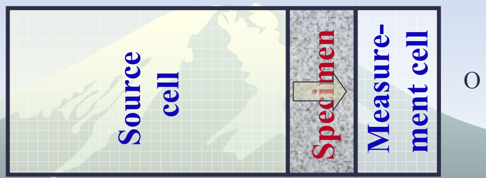
3) Theoretically evaluate applicability & limitations of conventional through-diffusion test

4) Show an example of improved technique

Concept of conventional through-diffusion test

Place a large cell at inlet side to dampen concentration variations

Replace the solution in measurement cell with fresh solution



Approximate solution: constant inlet & outlet concentrations

Effective diffusion coefficient: steady-state measurements

Potential problems: Time-consuming & cumbersome

Difference between analysis assumptions & test conditions

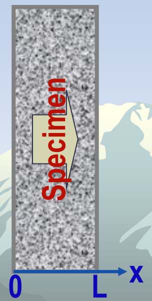
Rigorous solutions to through-diffusion test
(Mathematical models)

Boundary conditions

Source cell

Conventional & improved

$c(0,t) = c_1$



Measurement cell

Conventional

$c(L,t) = 0$

Improved

$$V_d \cdot \left.\frac{\partial c}{\partial t}\right|_{x=L} + A \cdot De \cdot \left.\frac{\partial c}{\partial x}\right|_{x=L} = 0$$

Initial condition $c(x,0) = 0$

Governing Equation

$$\frac{\partial c}{\partial t} = \frac{D_e}{\alpha}\frac{\partial^2 c}{\partial x^2}$$

$$\alpha = \varepsilon_{tot} + K_d \rho$$

c: Concentration; t: Time; De: Effective diffusion coefficient; α: Rock capacity factor; x: Distance; C1: Const. concentration in source cell; L: Length; Vd: Measurement cell volume; A: Cross-sectional area; ε_{tot}: total porosity; Kd : sorption coefficient; ρ : density

Approximate solution to through-diffusion test (Conventional)

Solution

$$c(x,t) = c_1\left[1 - \frac{x}{L} - \frac{2}{\pi}\sum_{n=1}^{\infty}\frac{1}{n}\cdot\sin\left(n\cdot\pi\cdot\frac{x}{L}\right)\cdot\exp\left(-\frac{n^2\cdot\pi^2\cdot D_e}{\alpha\cdot L^2}\cdot t\right)\right]$$

Total quantity diffused into measurement cell after time t

$$Q(t) = -D_e\cdot A\int_0^t \left.\frac{\partial(x,\tau)}{\partial x}\right|_{x=L} d\tau$$

$$= A\cdot L\cdot c_1\left\{\frac{D_e}{L^2}\cdot t - \frac{\alpha}{6} - \frac{2\alpha}{\pi^2}\sum_{n=1}^{\infty}\frac{(-1)^n}{n}\exp\left(-\frac{n^2\cdot\pi^2\cdot D_e}{\alpha\cdot L^2}\cdot t\right)\right\}$$

At long times or steady state

$$\left.Q(t)\right|_{t\to\infty} = A\cdot L\cdot c_1\left[\frac{D_e}{L^2}\cdot t - \frac{\alpha}{6}\right]$$

Time-lag method

Slope: $(A\cdot c_1\cdot D_e)/L$

Intercept on t axis: $(\alpha\cdot L^2)/(6D_e)$

Rigorous solution to through-diffusion test
(Improved, CIC-IOC)

Solution

$$c(x,t) = c_1\left\{1 - 2\sum_{m=0}^{\infty}\frac{(\lambda_m^2+\beta^2)\cdot\exp\left(-\frac{D_e\cdot\lambda_m^2}{\alpha\cdot L^2}\cdot t\right)\cdot\sin\left(\lambda_m\cdot\frac{x}{L}\right)}{\lambda_m\cdot\left[\lambda_m^2+\beta(\beta+1)\right]}\right\}$$

$$\beta = (\alpha\cdot A\cdot L)/V_d \qquad \tan(\lambda) = \frac{\beta}{\lambda}$$

Total quantity diffused into measurement cell after time t

$$Q(t) = -D_e\cdot A\int_0^t \frac{\partial(x,\tau)}{\partial x}\Big|_{x=L} d\tau$$

$$= 2\alpha\cdot A\cdot L\cdot c_1\left\{\sum_{m=0}^{\infty}\frac{(\lambda_m^2+\beta^2)\cdot\cos(\lambda_m)}{\lambda_m^2\left[\lambda_m^2+\beta(\beta+1)\right]}\cdot\left[1-\exp\left(-\frac{D_e\cdot\lambda_m^2}{\alpha\cdot L^2}\cdot t\right)\right]\right\}$$

At steady state: c(x, t)=c1; Q(t)->constant; No more diffusion

Theoretical evaluation
(Approach)

1) Numerically producing "experimental data" using the rigorous solution

2) Back-calculating effective diffusion coefficient & rock capacity factor using the conventional time-lag method

3) Comparing the back-calculated & input values for the two parameters

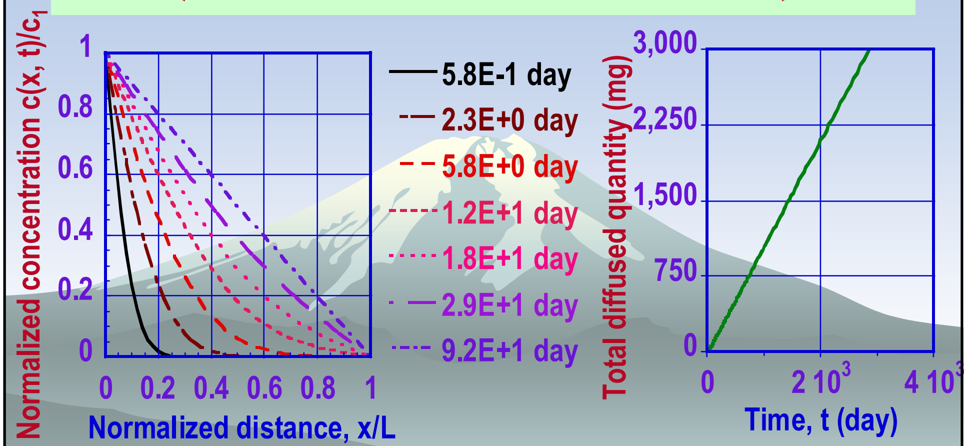
Theoretical evaluation
(Hypothetical test conditions)

Hypothetical test conditions for theoretical simulations

Specimen 	Cross-sectional area (m2) Length (m) Effective diffusion coefficient (m2/s) Rock capacity factor	3.85E-3 1.00E-2 2.5E-13 3.5E-2
Source cell	Concentration (ppm)	127000
Measurement cell	Volume (m3)	4.00E-5

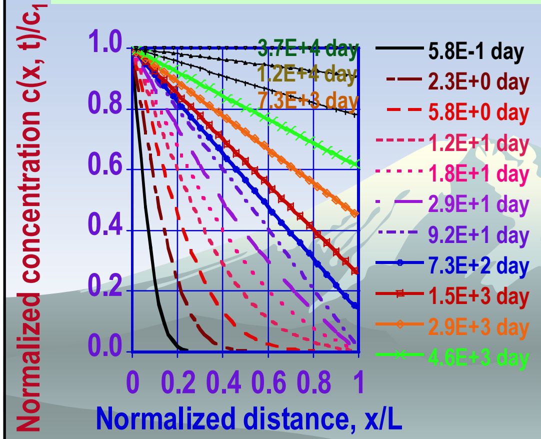
Specimen dimensions: our on-going experiments
Other parameters: test results reported in a number of articles

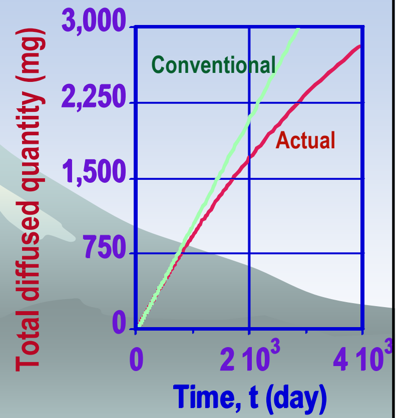
Theoretical evaluation
(Simulated results of conventional test)



At onset of experiment, concentration at inlet end is suddenly increased
Concentrations along the specimen axis gradually increase
Finally reaching a linear distribution, i.e., a steady-state condition
Total diffused quantity increases linearly

Theoretical evaluation
(Simulated results of improved test, CIC-IOC)





At onset of experiment, concentration at inlet end is suddenly increased
Concentrations along the specimen axis gradually increase
Finally reaching the concentration of source solution
Total diffused quantity converges to a certain value

Theoretical evaluation
(Errors from time-lag method)

Definition of elative errors:

$$D_{e-err} = \frac{D_{e-determined}}{D_{e-input}} \cdot 100\% \qquad \alpha_{err} = \frac{\alpha_{determined}}{\alpha_{input}} \cdot 100\%$$

Relative errors in determining effective diffusion coefficient & rock capacity factor by using the time-lag method

Relative errors	Concentration increase in measurement cell (% of constant concentration in source cell)			
	1%	3%	~~3%~~ 5%	9%
D_{e-err} (%)	96	96	94	88
α_{err} (%)	93	90	73	26

Conventional through-diffusion test based on time-lag method tends to underestimate both the two parameters

Another improvement to through-diffusion test

(Decreasing inlet concentration-Increasing outlet concentration)

Boundary conditions

Source cell

$c(0,0)=c_1$

$$V_u \cdot \left.\frac{\partial c}{\partial t}\right|_{x=0} - A \cdot De \cdot \left.\frac{\partial c}{\partial x}\right|_{x=0} = 0$$

Measurement cell

$c(L,0)=0$

$$V_d \cdot \left.\frac{\partial c}{\partial t}\right|_{x=L} + A \cdot De \cdot \left.\frac{\partial c}{\partial x}\right|_{x=L} = 0$$

Rigorous solution:

Initial condition $c(x,0)=0$

$$c(x,t)=\frac{c_1}{\delta+\gamma+1}-2c_1\sum_{m=0}^{\infty}\frac{\exp\left(-\frac{D_e\cdot\phi_m^2}{\alpha\cdot L^2}\cdot t\right)\cdot\left[\delta\cdot\cos\left(\phi_m\cdot\frac{L-x}{L}\right)-\gamma\cdot\phi_m\cdot\sin\left(\phi_m\cdot\frac{L-x}{L}\right)\right]}{\left[\gamma\cdot\phi_m^2-\delta(\delta+\gamma+1)\right]\cos(\phi_m)+\left[\delta\cdot\gamma+\delta+2\gamma\right]\phi_m\cdot\sin(\phi_m)}$$

$\delta=(\alpha\cdot A\cdot L)/V_u$ $\qquad$ $\gamma=V_d/V_u$ $\qquad$ $\tan(\phi)=\frac{\delta\cdot(\gamma+1)\cdot\phi}{\gamma\cdot\phi^2-\delta^2}$

Technique for interpreting test data

(Parameter identification technique)

Solutions are rigorous; Difficult to determine analytically.
Graphical methods possible; Procedures cumbersome; Accuracy limited.

Error function:
(for CIC-IOC method)

$$\varepsilon=\sum_{i=1}^{n}\left\{Q(t_i)-Q(t_i)^{*}_{(D_e,\alpha)}\right\}^2$$

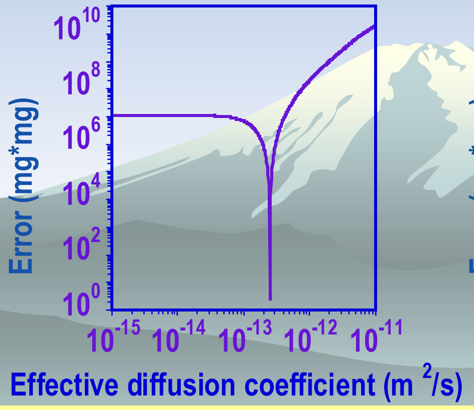
A least-squares reduction of discrepancy between measured and theoretically calculated results

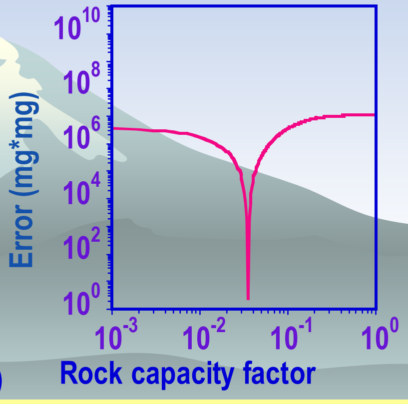
N: the number of measured data points

Technique for interpreting test data

(Characteristics of the error function, CIC-IOC)

Varying one parameter across a certain span of magnitude
Maintaining the other parameter constant & equal to its "real value"





Error curves are convex in the neighboring domains of the "real values"
It is possible to uniquely quantify the magnitudes of the parameters

PARAMETER BACK-CALCULATION TECHNIQUE

(Decreasing inlet concentration-Increasing outlet concentration)

Error function

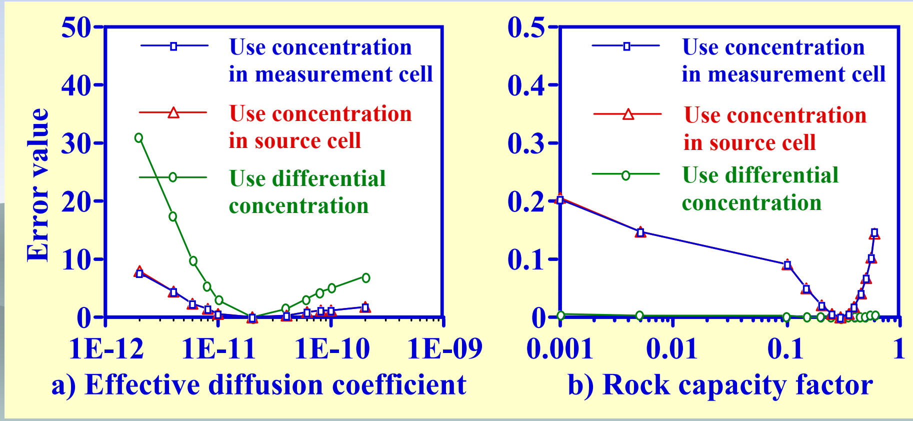
$$\varepsilon = \sum_{i=1}^{n} \left\{ c(t_i)_{(D_{e,\alpha})} - c(t_i)^{*}_{(D_{e,\alpha})} \right\}^2$$

n is the number of measured data points; $c(t_i)$ and $c(t_i)^*$ are the concentrations measured at time t_i

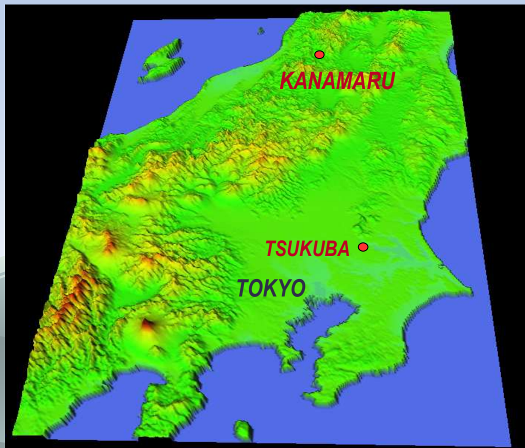
Hypothetical test conditions for the theoretical simulations

Specimen dimension	Diameter (m) Equivalent cross sectional area (m^2) Length (m)	0.064 3.22E-3 0.01
Transpor properties	Effective diffusion coefficient (m^2/s) Rock capacity factor	2E-11 0.15
Initial concentration	Normalized concentration	1
Volumes of cells	Source cell (m^3) Measurement cell (m^3)	4.4E-5 4.4E-5

Characteristics of the error function
(Decreasing inlet concentration-Increasing outlet concentration)



Location of Kanamaru research site



The specimen was taken from a depth of 12.03 to 12.04m below ground level. At about this depth, a several centimeters thick, poor quality uranium deposit has been detected. The diameter of the specimen was 64mm.

Test conditions

a) Front view b) Side view c) Oblique view

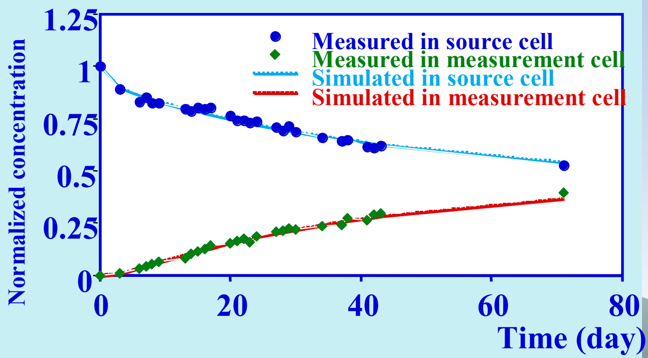
The volume of cells was 44 ml.

Non-sorbing ion Cl^- was chosen as a tracer (NaCl) and its concentrations were measured using Ion Chromatography. 20 micro-liters of solution was taken for each sampling.

The final pH values : 7.37 and 7.33 respectively.

Temperature controlled room (22Centigrade).

Test results



Transport property (Tracer ion, Cl^-)	Data used for the analyses		
	Concentration decrease in source cell	Concentration increase in measurement cell	Differential concentration between the two
Effective diffusion coefficient (m^2/s)	1.80E-11	2.03E-11	1.98E-11
Rock capacity factor	0.38	0.30	0.34

Note: Rock type: Sedimentary rock; Specimen number: M1-S-2A

Concluding remarks (1)

Laboratory diffusion test:

Well-established & widely adopted approach for characterizing the transport properties of geo-materials

Conventional through-diffusion tests:

May be time-consuming, cumbersome

May cause errors in the effective diffusion coefficient & rock capacity factor due to the difference between actual test conditions & analytical assumptions

Concluding remarks (2)

If solution in measurement cell is not replaced with fresh solution to maintain 0 concentration condition &
data are interpreted with conventional time-lag method

There will be a tendency to underestimate both the effective diffusion coefficient & rock capacity factor

The higher the concentration increase in the measurement cell, the larger will be the error in estimating the two parameters

Concluding remarks (3)

The effective diffusion coefficient & rock capacity factor can be uniquely back-calculated from the measurements of the improved (i.e., CIC-IOC and DIC-IOC) diffusion tests

Rigorous theoretical solution

+

Parameter identification technique

Concluding remarks (4)

The improved technique enables the use of the data continuously measured in the measurement cell

May allow automation of laboratory diffusion tests if the concentrations can be automatically detected by appropriate sensors

Test management may become easier & test efficiency may be highly increased

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To end this talk…

Accurate determination of parameters related to mass transport assessment is very important

Traditional test methods, although considered to be well-established and are widely adopted in engineering practices, does not necessarily provide accurate results if their applicable conditions are not well confirmed

Considerations and approaches presented in this talk may offer basic ideas to avoid misinterpreting the experimental data

Thank you very much for your kind attention!