Evaluation of long-term durability of engineered barrier system (EBS) of bentonite and cementitious materials by migration technique

K. Nakarai*, M. Watanabe*, T. Sugiyama**, Y. Tsuji*

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Contents of this presentation

1. **Introduction**
   - Concept of EBS for LLW in Japan
   - Degradation of EBS at cement/bentonite interaction
2. **Test method**
   - Acceleration test by electrical migration technique
3. **Experimental procedures and results**
   1. Investigation of effect of dry density on degradation of EBS at cement/bentonite interaction
   2. Investigation of effect mixed $NaHCO_3$ on degradation of EBS at cement/bentonite interaction
4. **Conclusions**

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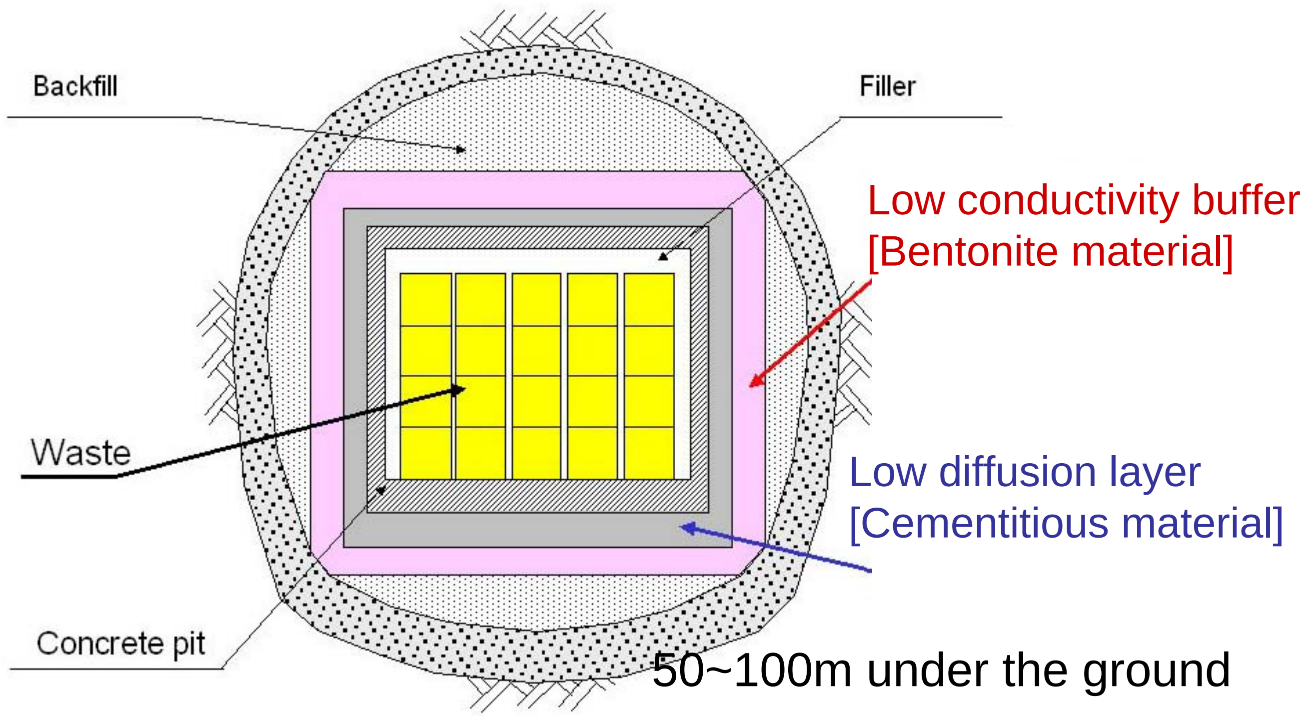
EBS for LLW in Japan

Underground cavern type disposal facility to isolate low-level radioactive waste

(http://www.enecho.meti.go.jp/rw/gaiyo/gaiyo03-3.html)



Repository concept of LLW in Japan

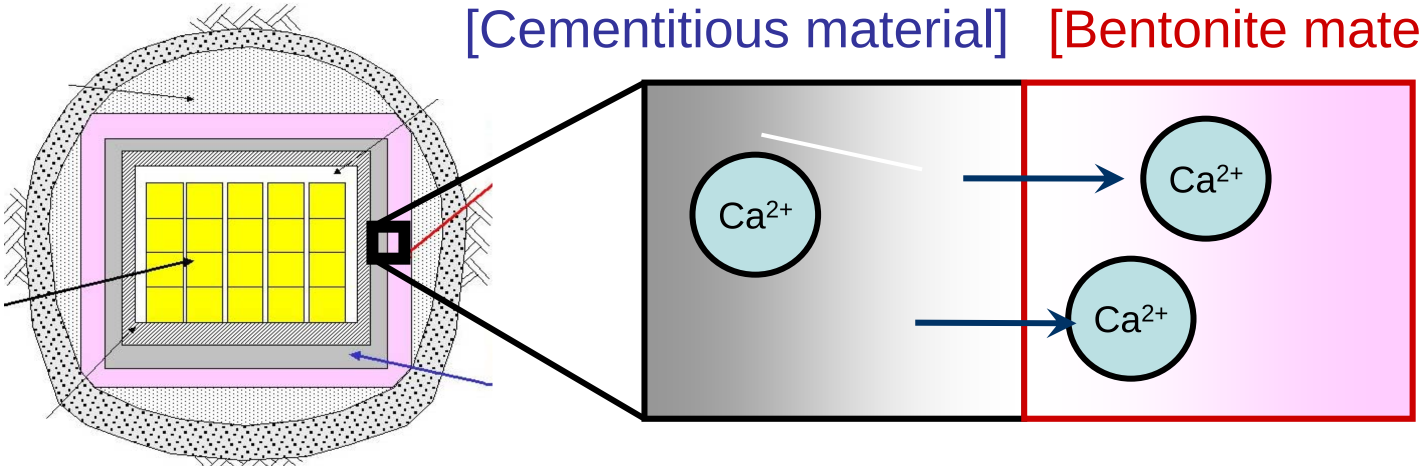
Extremely long-term stability for several tens thousands years is required.

Durability problem of EBS for LLW in Japan

To evaluate extremely long-term stability during tens thousands of years

Low diffusion layer
[Cementitious material]

Low conductivity buffer
[Bentonite material]



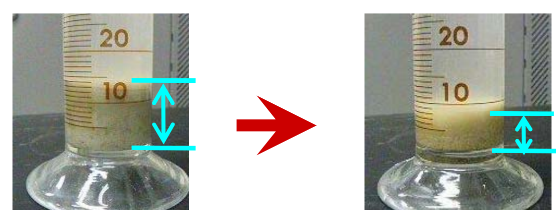
Repository concept
of LLW in Japan

Ca leaching → Increase in Ca & pH

Degradation
Increase in porosity
>>>Increase in diffusivity

Degradation
Decrease in swelling capacity
>>>Increase in conductivity

Na
-type

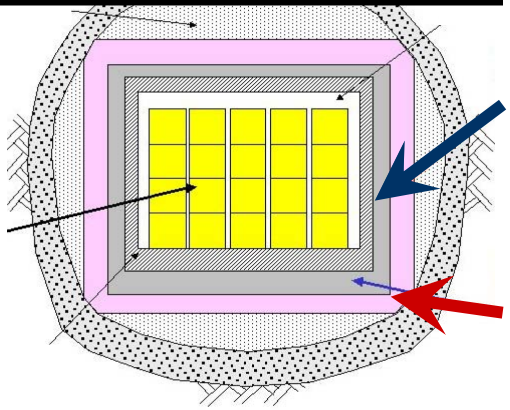


Ca
-type

Two experimental plans in this study

Problem of EBS

To evaluate stability during tens thousands of years



Low diffusion layer
[Cementitious material]

Low conductivity buffer
[Bentonite material]

Leaching → Degradation
Ex) Increase in diffusivity

↕

Increase in Ca & pH → Degradation
Ex) Increase in conductivity

Repository concept of LLW in Japan

Approach I

Reduce negative cement/bentonite interaction

Exp I → Increase in dry density of bentonite

Approach II

Control cement/bentonite interaction for increasing stability

Exp II → Mixing $NaHCO_3$ for creating additional layer of $CaCO_3$

Next topic

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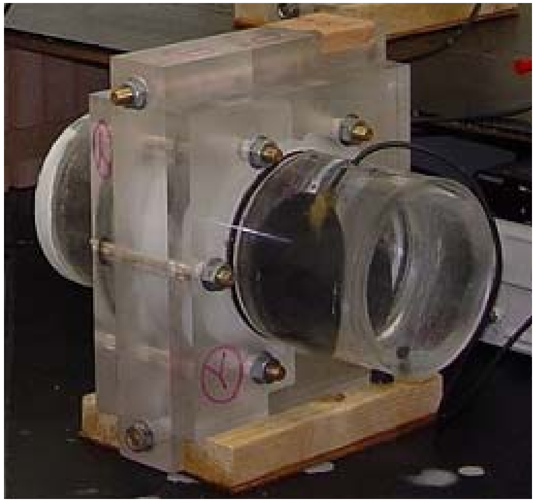
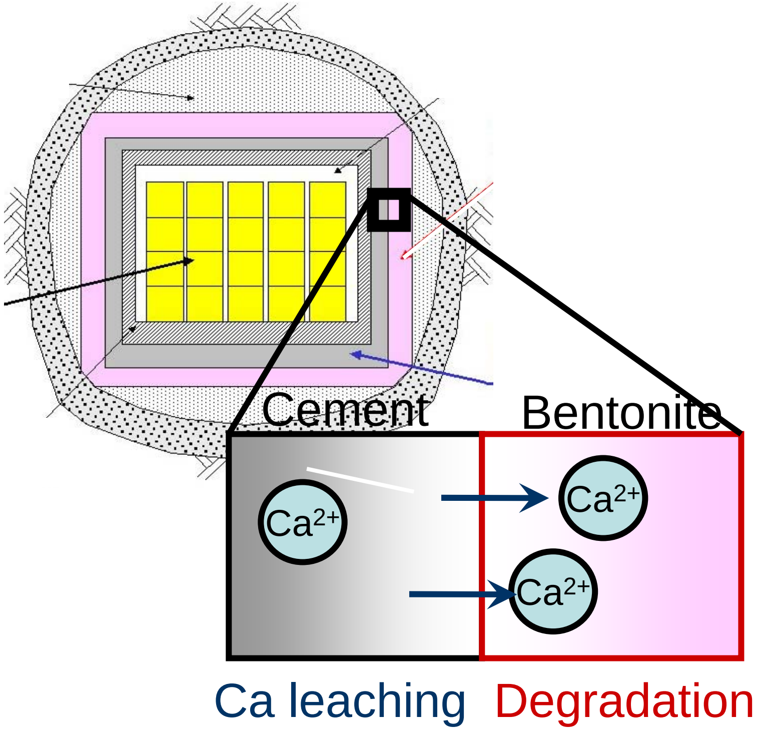
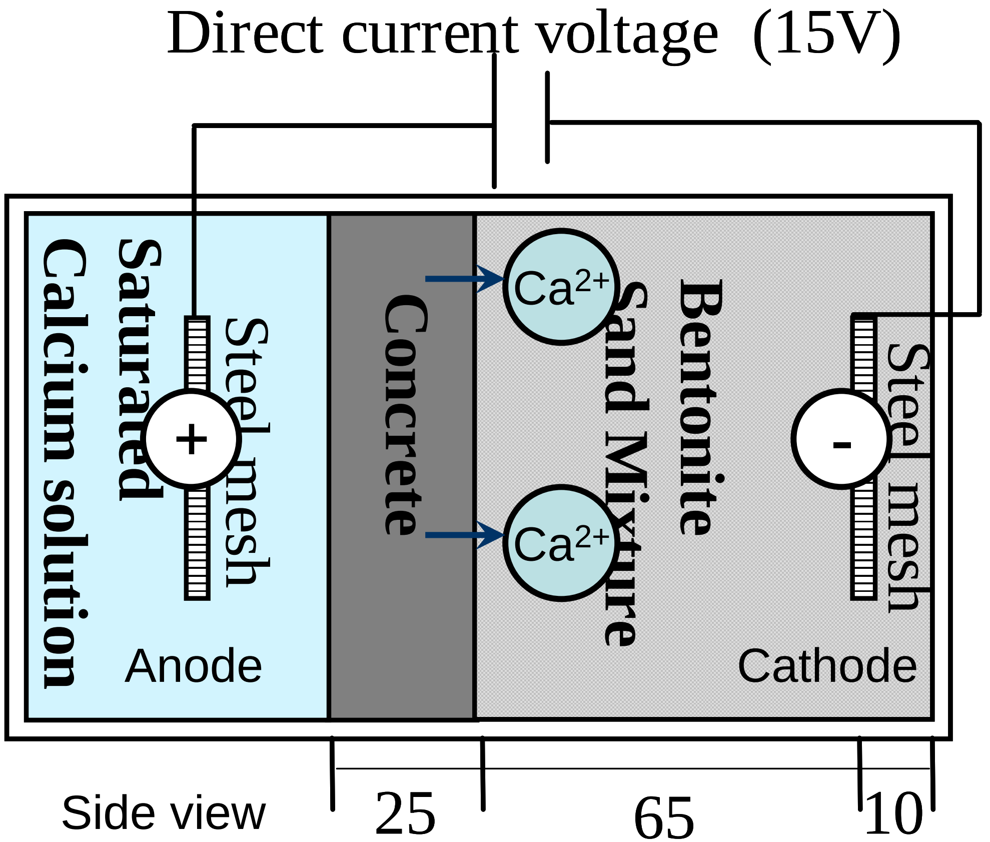
Acceleration test by electrical migration technique



To accelerate ion transport
by applying electric potential gradient

Several studies have been reported.
(e.g. Saito et al. 1997)





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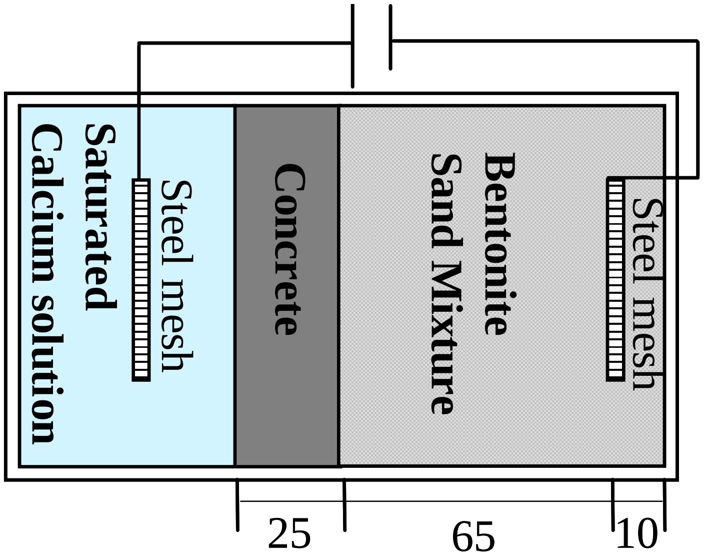
Exp I: Influence of dry density (Specimen)



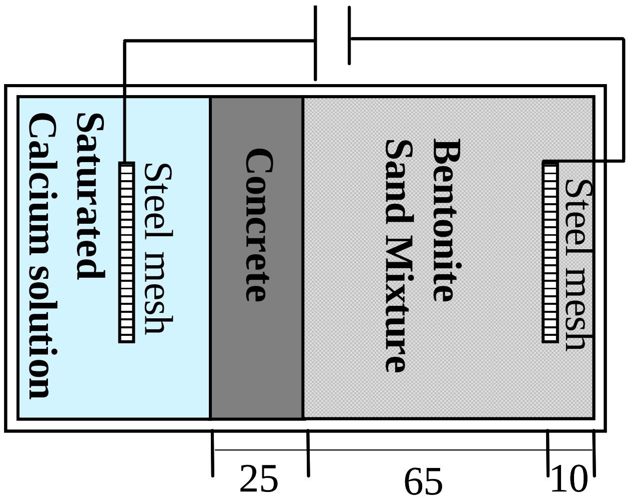
[Cementitious material]
Concrete
Cement: OPC
W/C = 55%

[Bentonite material]
Bentonite sand mixture
Bentonite: Kunigel V1
Bentonite:Sand = 7:3

4 specimens of bentonite sand mixture

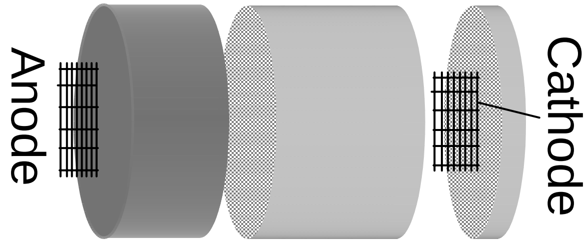
Name of specimen	Dry Density g/cm^3	Water Content %	Compaction times / Layer
Bt16	1.6	28.6	11
Bt17	1.7	26.1	19
Bt18	1.8	22.6	38
Bt19	1.9	15.0	150, 200

Exp I: Measurement after electrical migration test



No.	Dry density, g/cm^3	Water content, %
Bt16	1.6	28.6
Bt17	1.7	26.1
Bt18	1.8	22.6
Bt19	1.9	15.0

After 13.0kC accumulated electrical charge



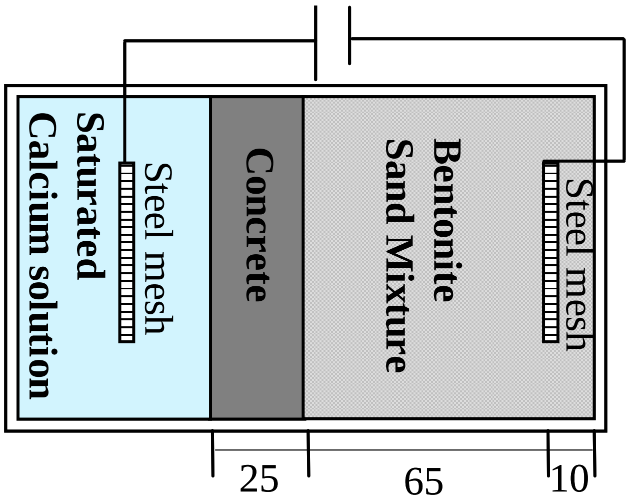
Concrete

TGA
Ca(OH)$_2$,

Swelling capacity
Cation concentration
EPMA (Bt19)

Bentonite sand mixture

Exp I: Measurement after electrical migration test



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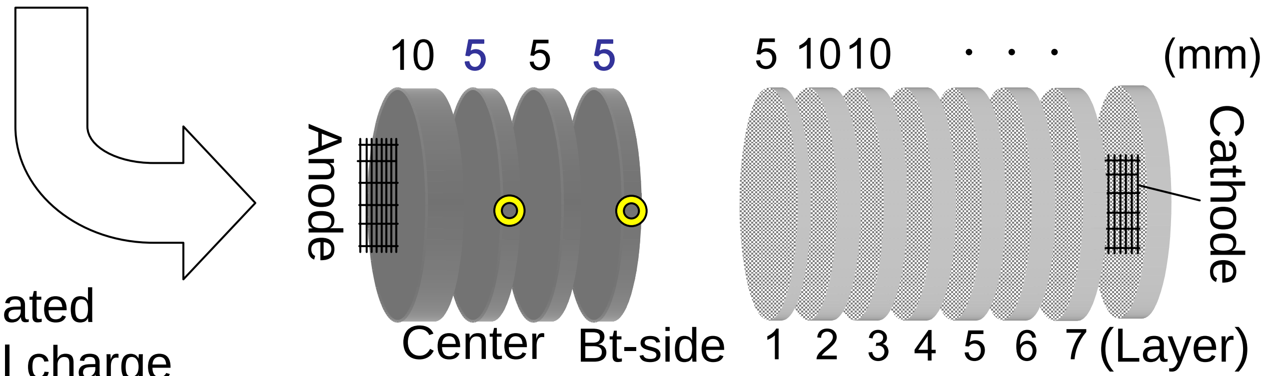


Concrete

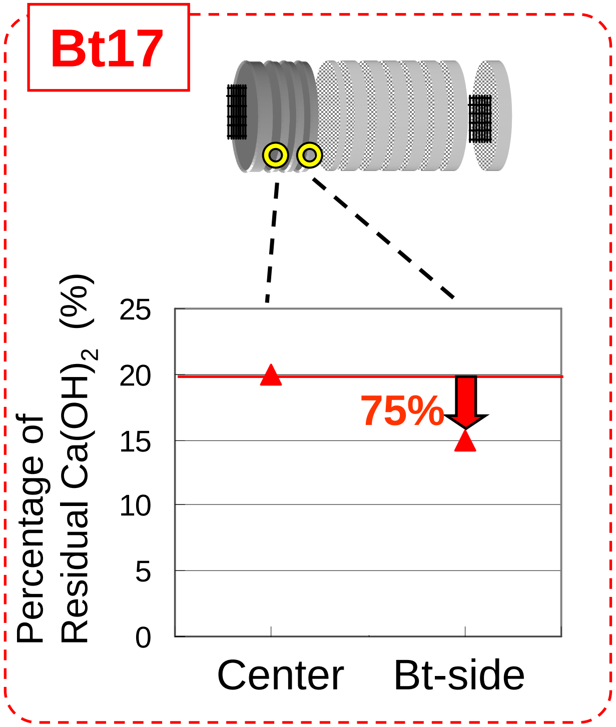
TGA
$Ca(OH)_2$,

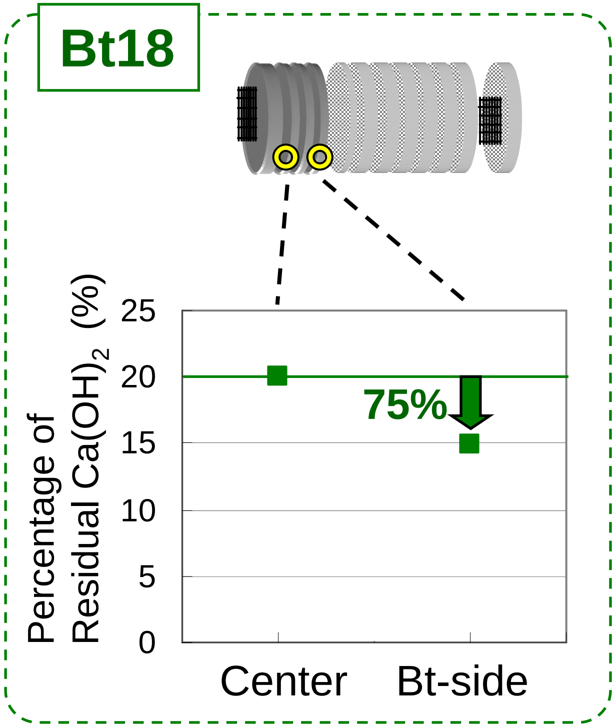
Swelling capacity
Cation concentration
EPMA (Bt19)

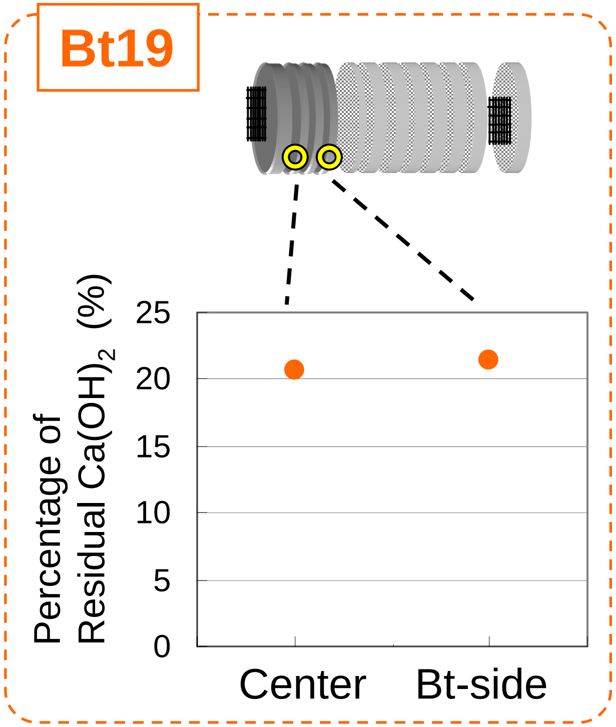
Bentonite sand mixture

Result of TGA: Degradation of concrete

$Ca(OH)_2$: leach from concrete first ⇨ Measurement of residual $Ca(OH)_2$







Bt17 **Bt18**

Percentage of residual $Ca(OH)_2$: **Center > Bt-side**

⇩

Calcium leached from Bt-side of concrete to bentonite

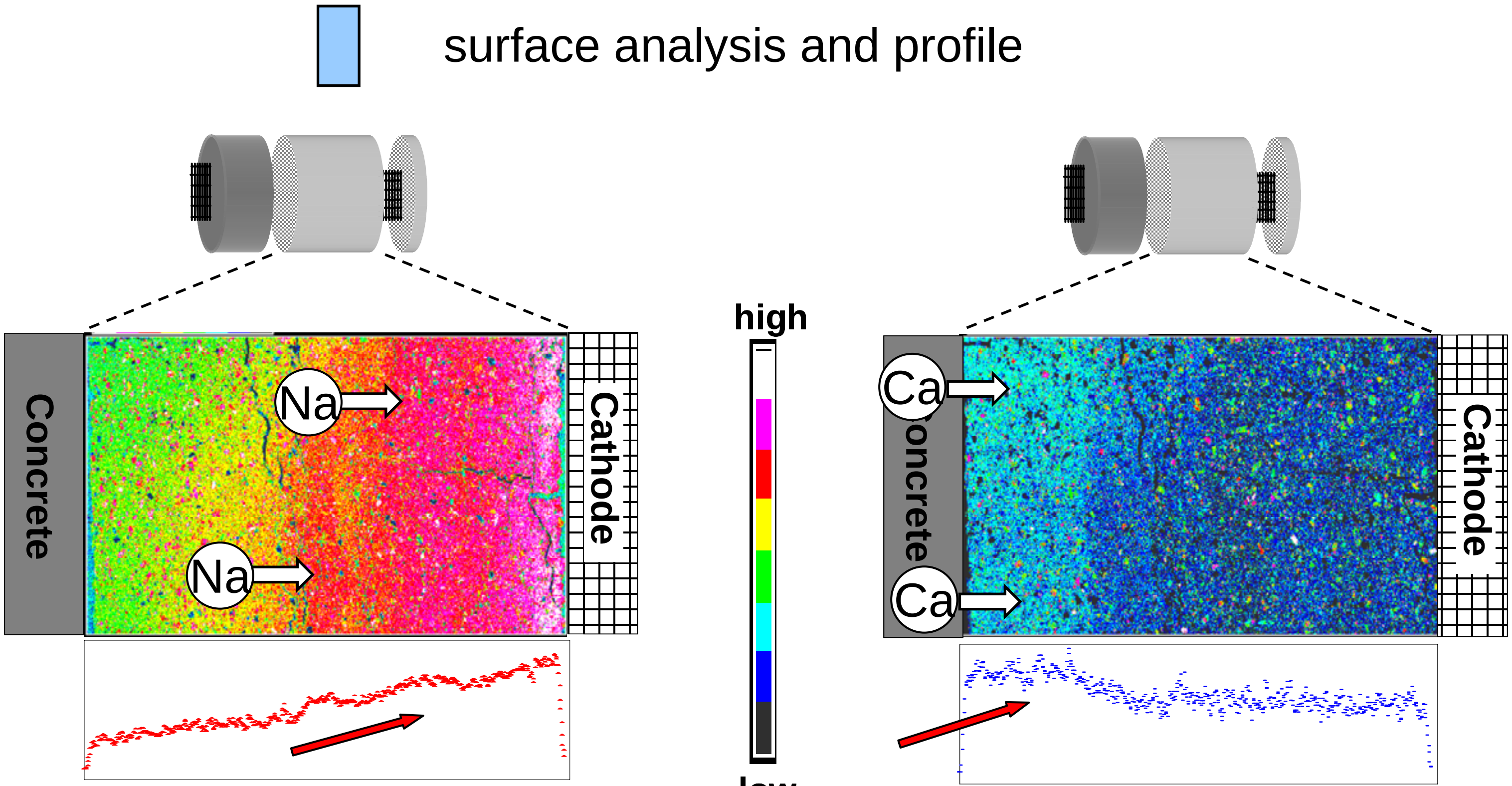
Bt19

Decrease in Bt-side was not observed

⇩

Reduce degradation

Result of EPMA: Cations in bentonite (Bt19)



Profile of Sodium

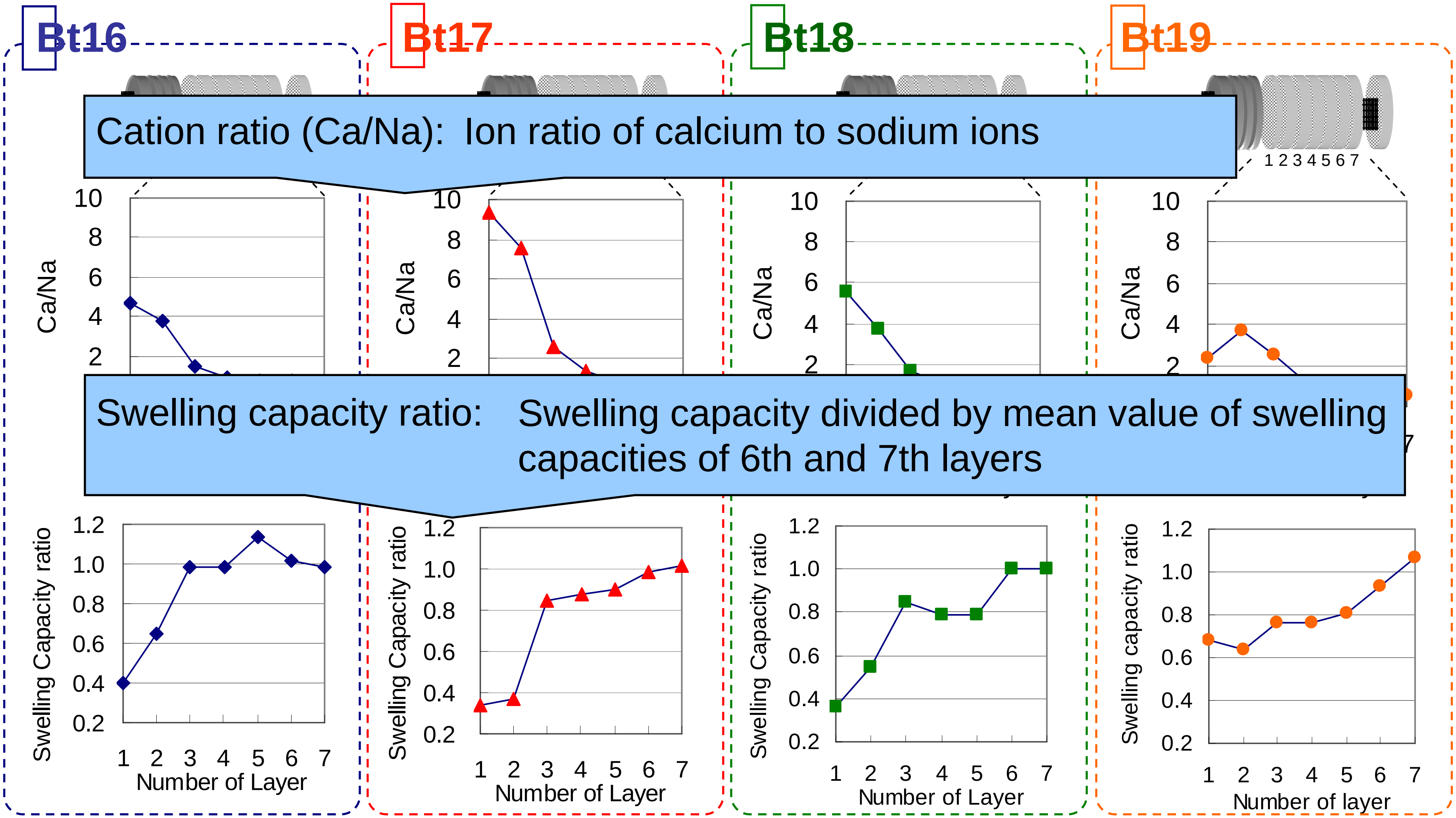
Profile of Calcium

Sodium Ion:
Migrated to cathode

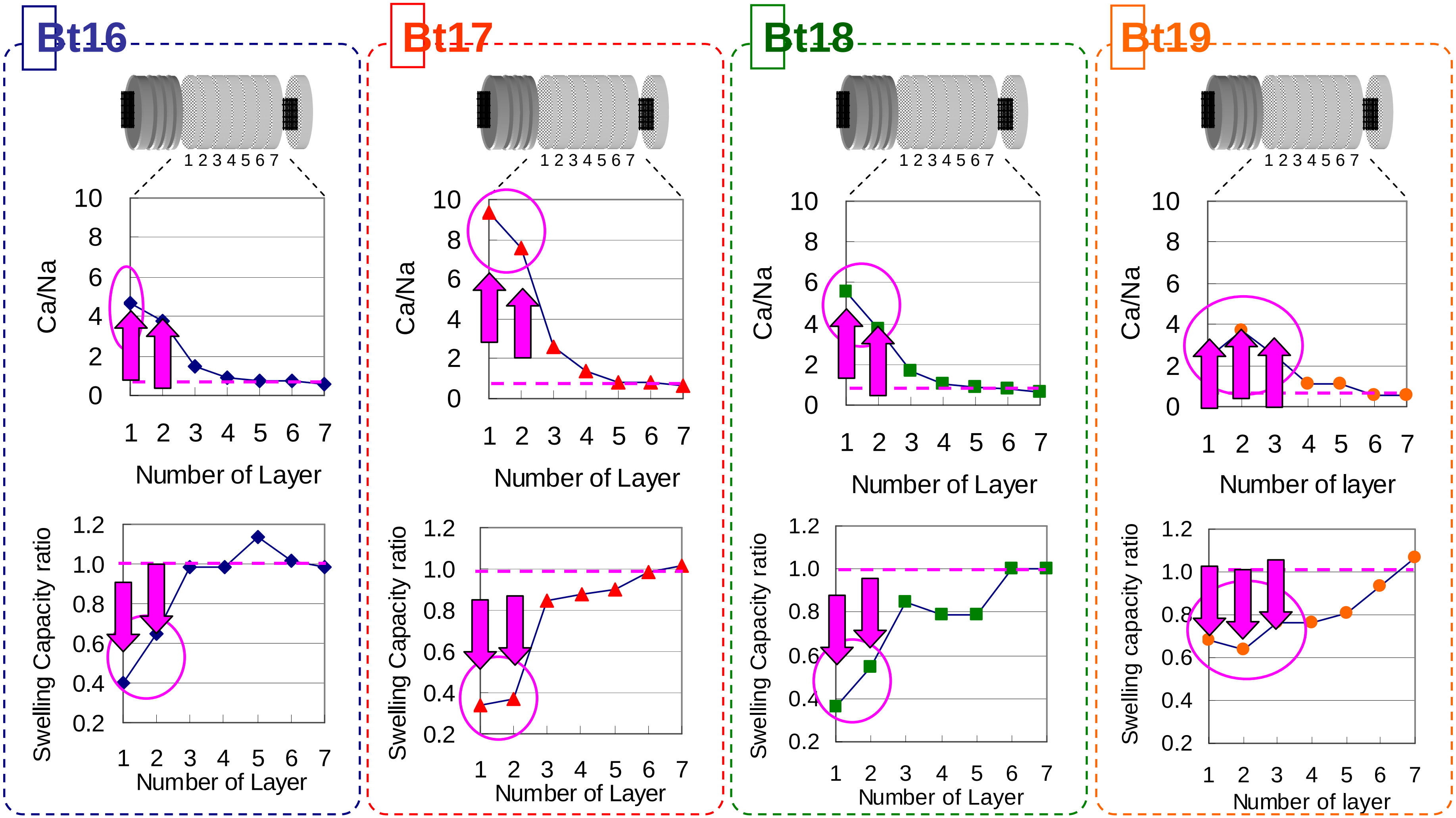
Calcium Ion:
Migrated from concrete

Alteration of bentonite progress from the interface

Result of cations and swelling capacity of bentonite



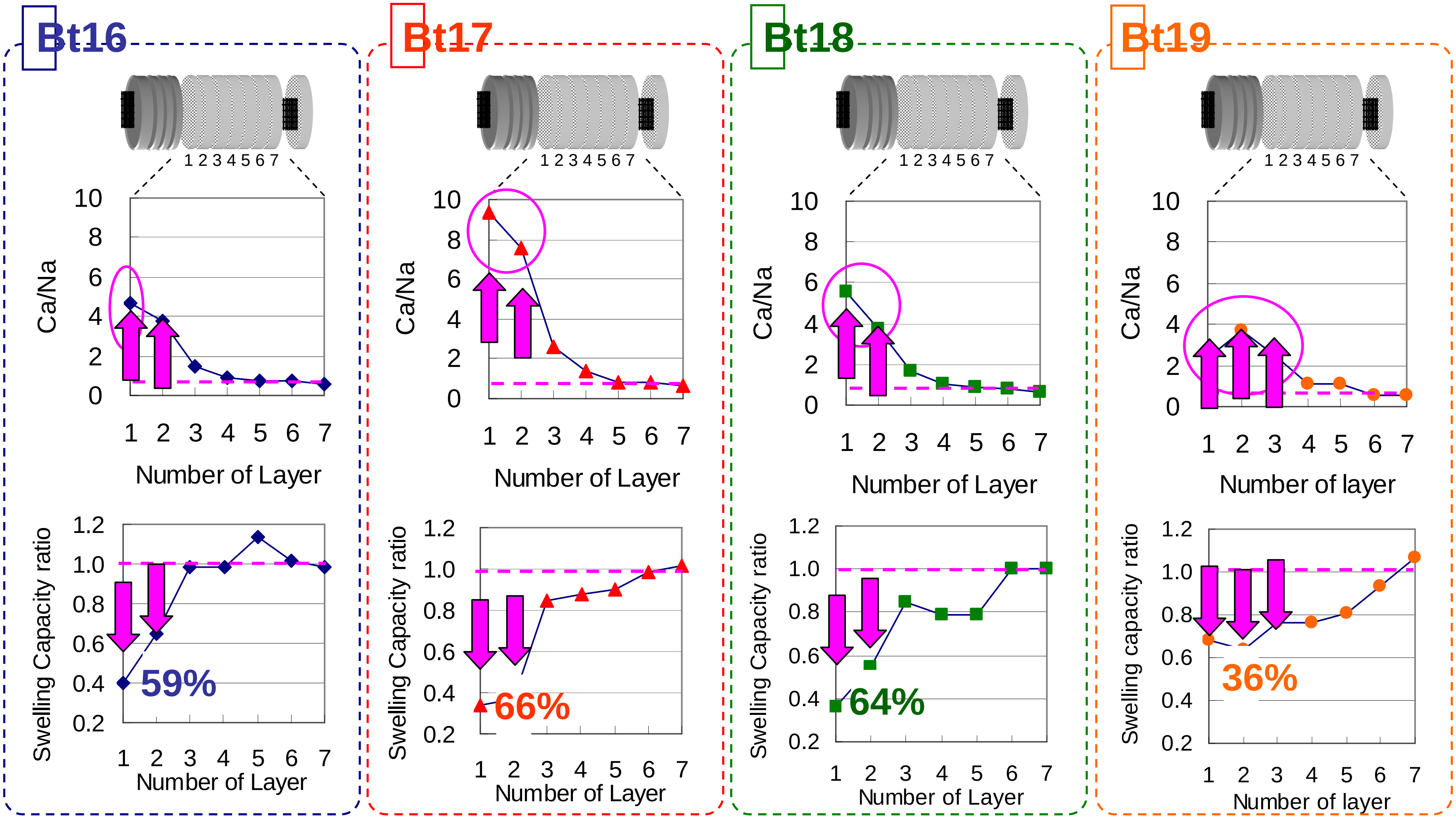
Result of cations and swelling capacity of bentonite



Concrete side:

Swelling capacities decrease with increase in Ca/Na ⇨ Change to Ca-type

Result of cations and swelling capacity of bentonite



Bt19: Decrease in swelling capacity was reduced ⇒ Low conductivity can be maintained by using high dry density bentonite

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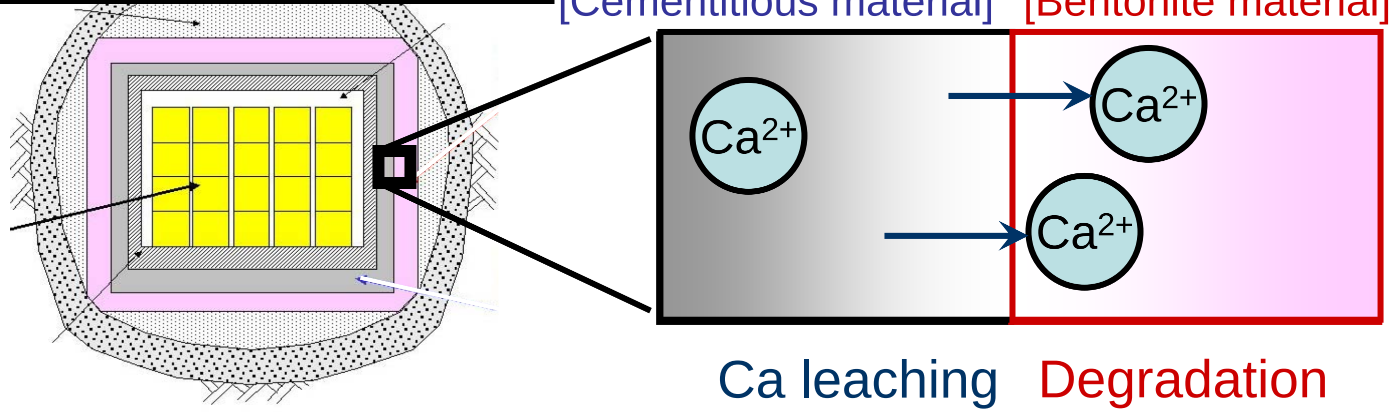
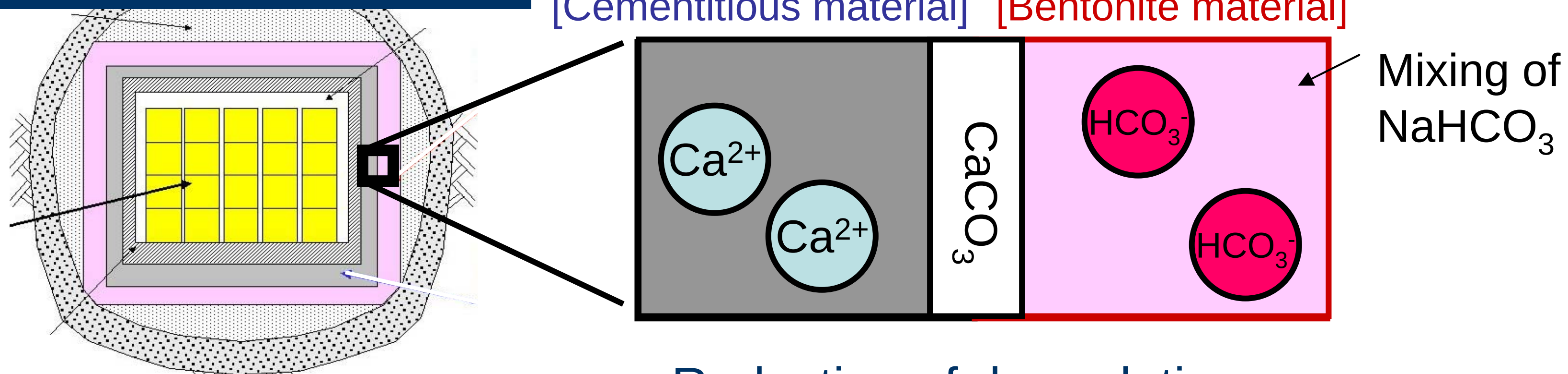
Exp II: Utilization of cement/bentonite interaction

Exp I: Negative effect



Exp II: Utilization



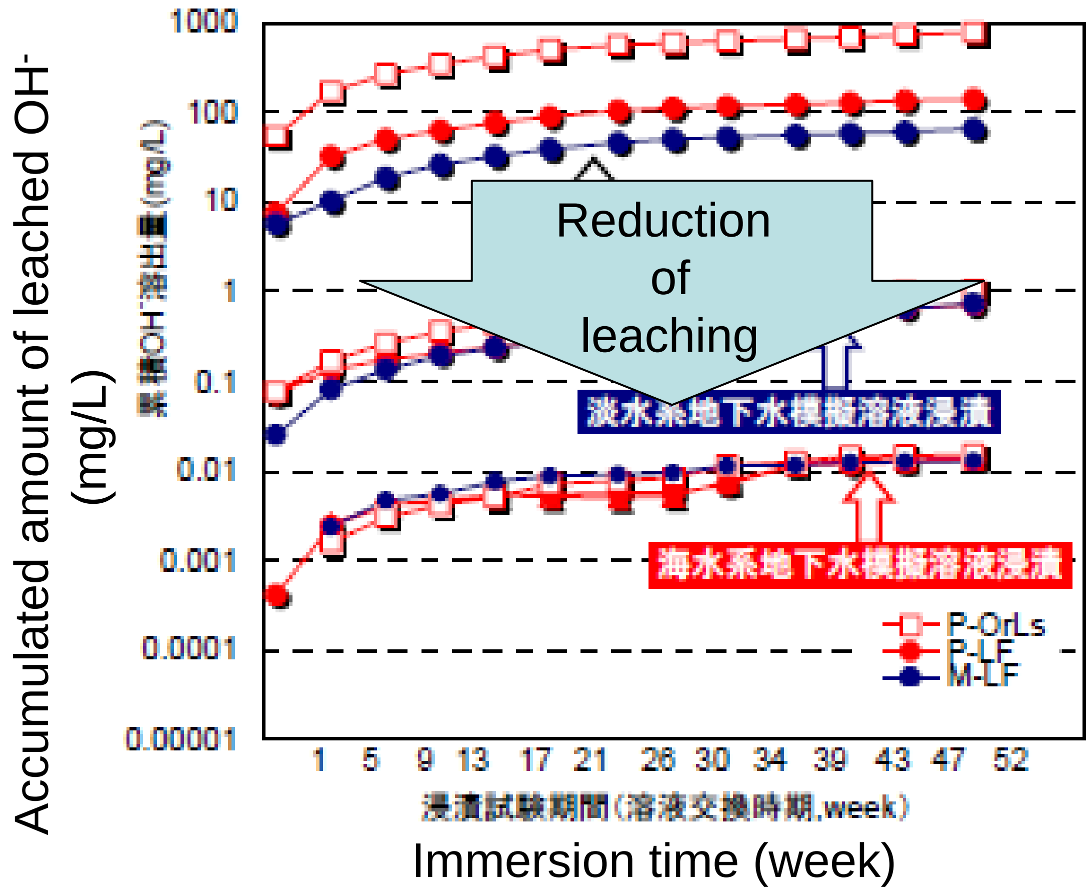
Mixing $NaHCO_3$ into bentonite

Reduction of degradation

Additional barrier of $CaCO_3$

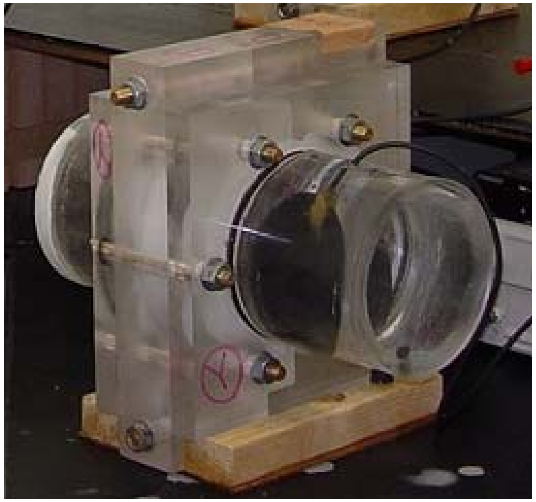
Reference: Reduce leaching by HCO_3^-

Kurashige et al. (2005) have shown that HCO_3^- in ground water reduce leaching of cementitious materials.



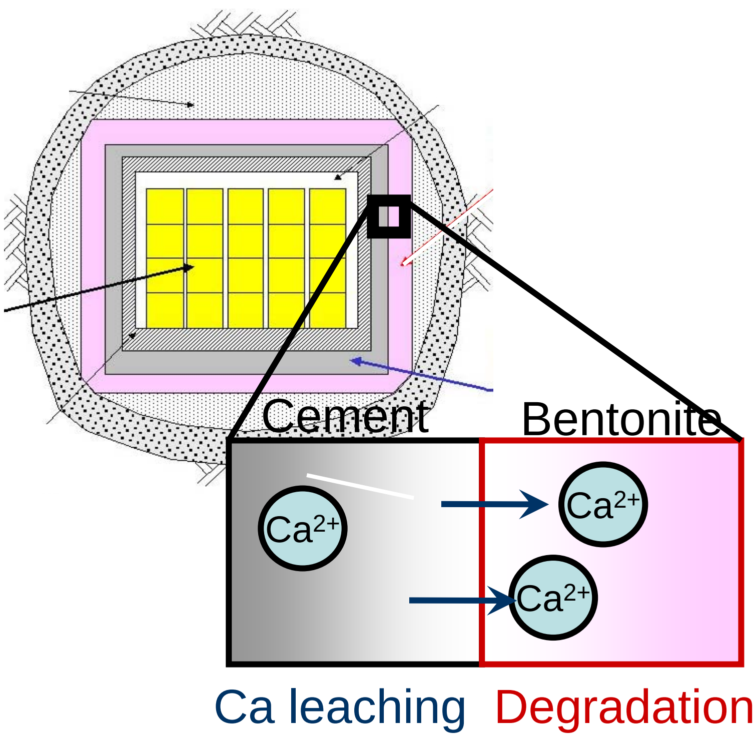
In this study, we aim to get this additional effect intentionally in the proposed artificial system.

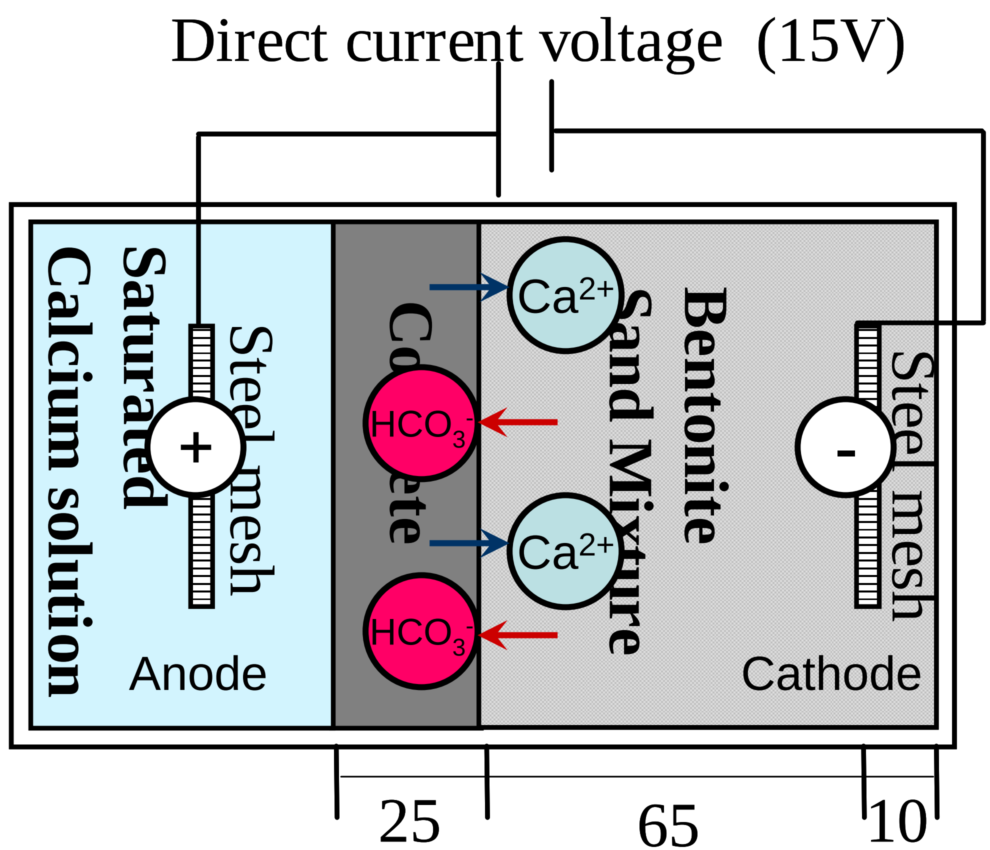
Exp II: Acceleration test by electrical migration technique



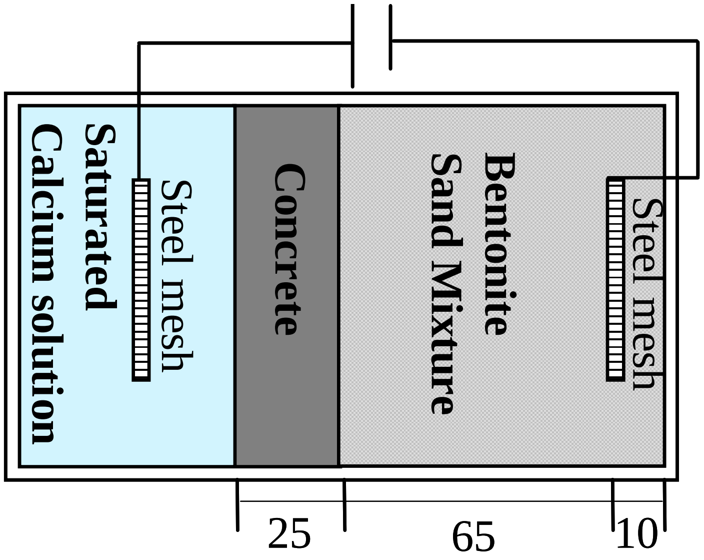
To accelerate ion transport
by applying electrical gradient

Ex) Saito et al. investigated calcium leaching





Exp II: Influence of mixing $NaHCO_3$ (Specimen)



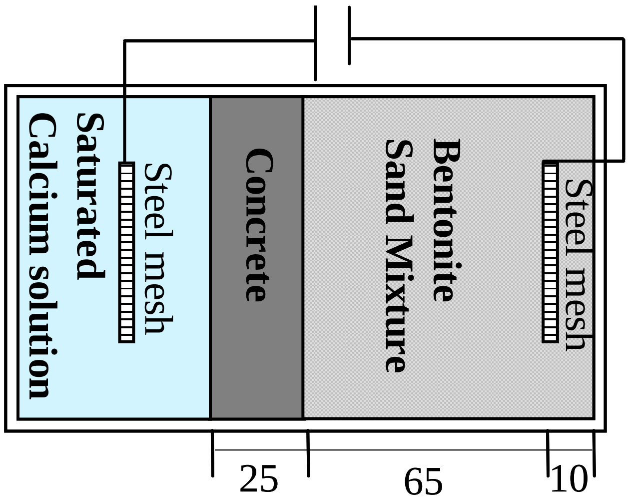
[Cementitious material]
Cement paste
Cement: OPC
W/C = 60%

[Bentonite material]
Bentonite sand mixture
Bentonite: Kunigel V1
Bentonite:Sand = 7:3
Dry density = 1.6 g/cm^3

4 specimens of bentonite sand mixture

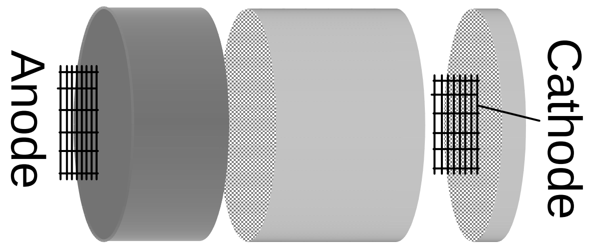
Name of specimen	$NaHCO_3$ mass %	Concentration g/litter	Remarks
C0	0	0	No mixing
C0.4	0.4	10	
C4	4.1	103	Saturation
C7	7.1	103	

Exp II: Measurement after electrical migration test



No.	$NaHCO_3$ mass %	Concentration g/litter
C0	0	0
C0.4	0.4	10
C4	4.1	103
C7	7.1	103

After
140 hours
electrical migration



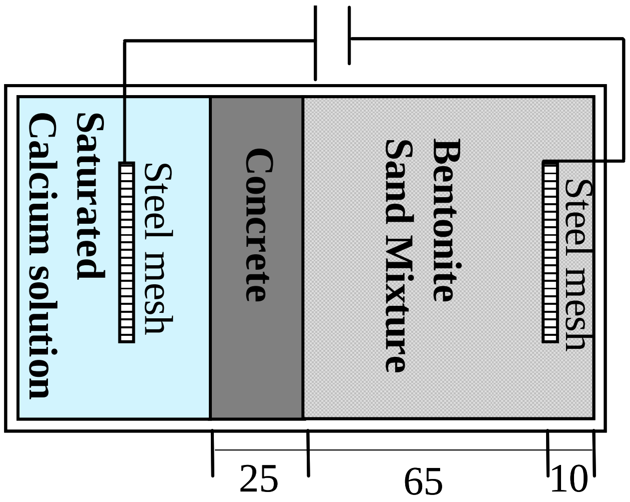
Cement paste

TGA
$Ca(OH)_2$, **$CaCO_3$**

Swelling capacity
Cation concentration

Bentonite sand mixture

Exp II: Measurement after electrical migration test



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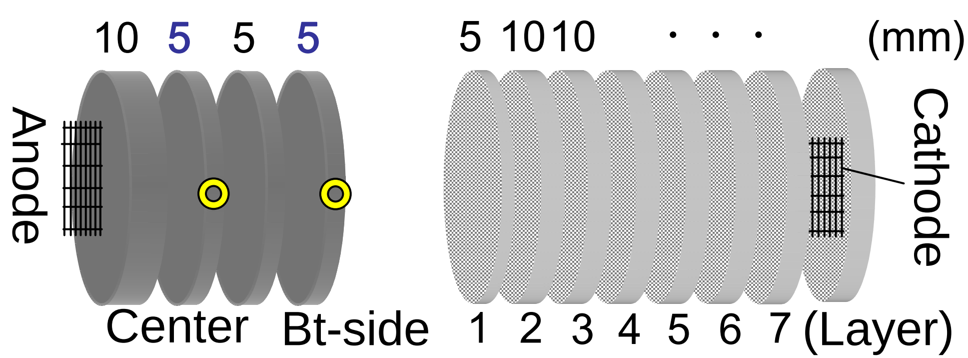


Cement paste

TGA
$Ca(OH)_2$, **$CaCO_3$**

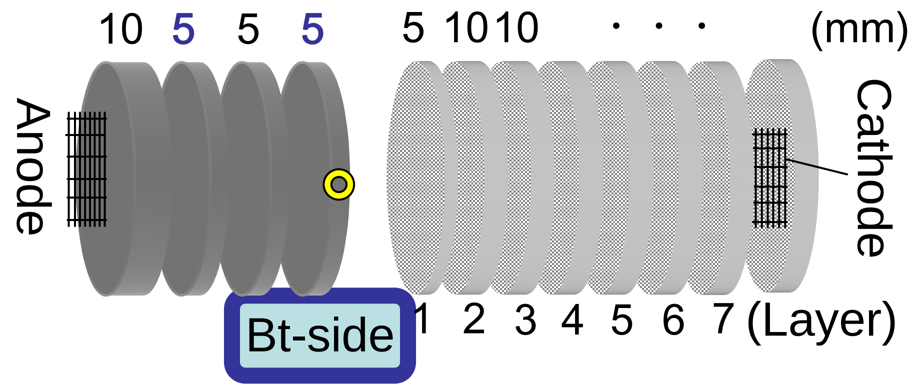
Swelling capacity
Cation concentration

Bentonite sand mixture

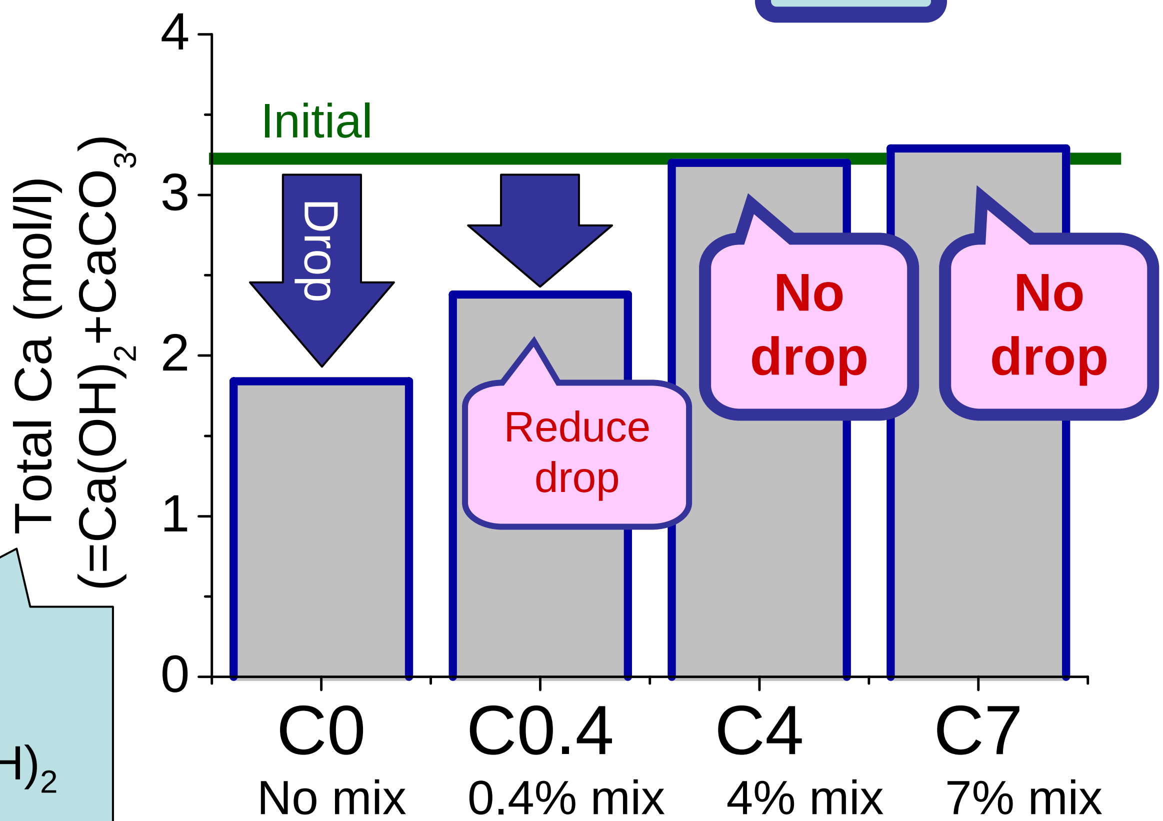
Result of TGA: Degradation of cement paste

Cement paste

TGA
→Measurement of $Ca(OH)_2$
→Measurement of $CaCO_3$



Total

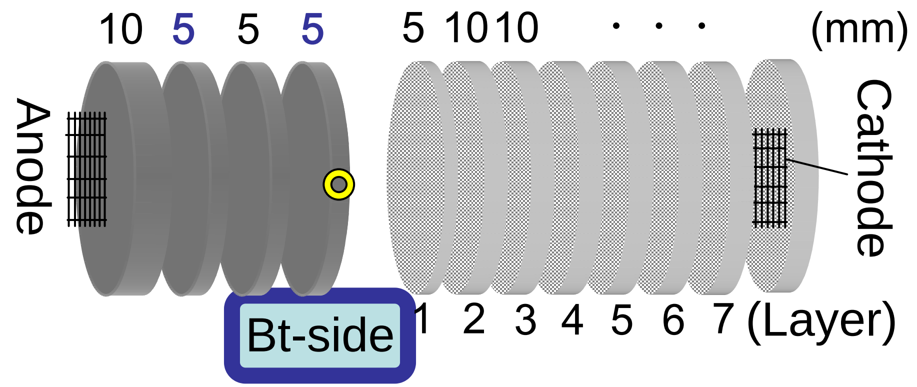


Calculated value as summation of Calcium in $Ca(OH)_2$ & $CaCO_3$

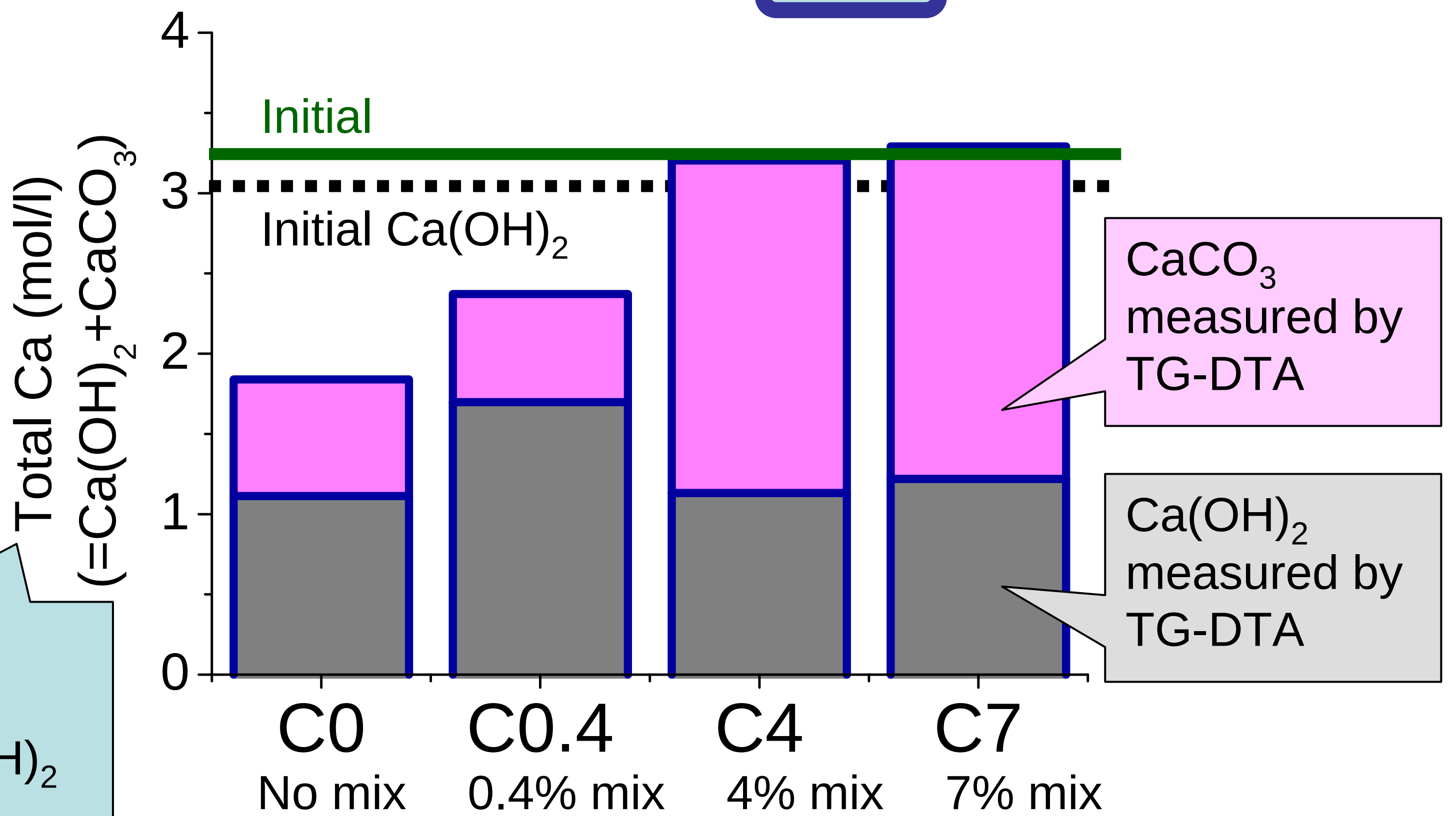
Result of TG-DTA: Degradation of cement paste

Cement paste

TG-DTA
→Measurement of $Ca(OH)_2$
→Measurement of $CaCO_3$



Details

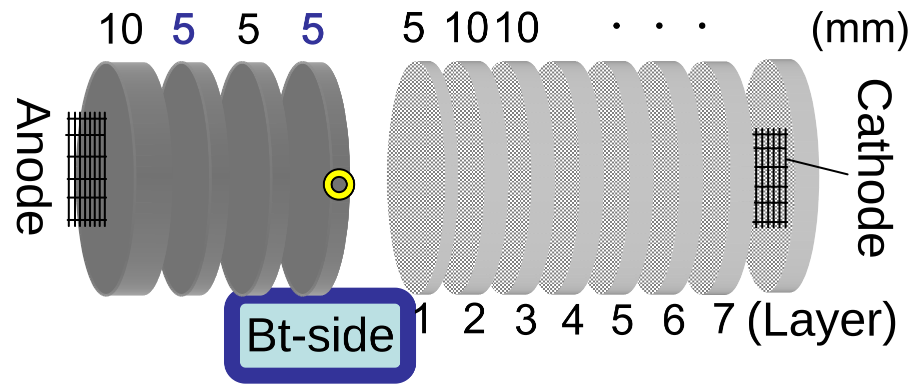


Calculated value as summation of Calcium in $Ca(OH)_2$ & $CaCO_3$

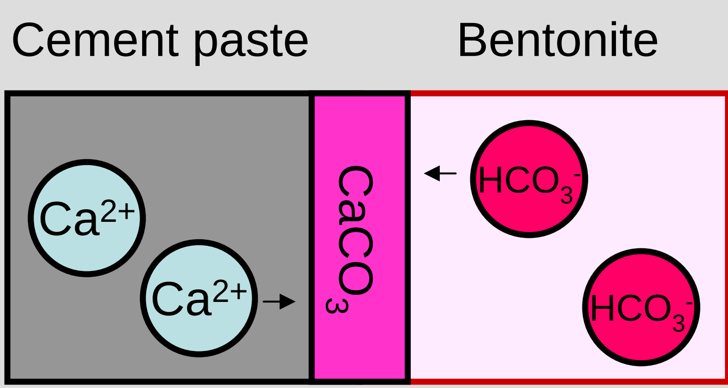
Result of TG-DTA: Degradation of cement paste

Cement paste

TG-DTA
→Measurement of $Ca(OH)_2$
→Measurement of $CaCO_3$

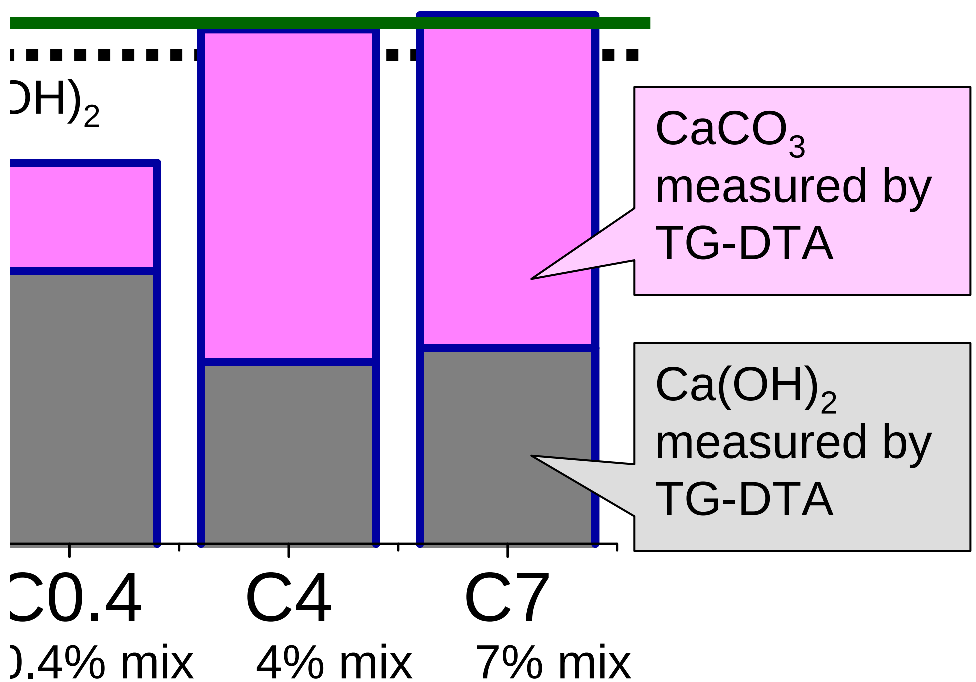


Detail



By mixing $NaHCO_3$ into bentonite,
$CaCO_3$ was precipitated
at cement/bentonite interface

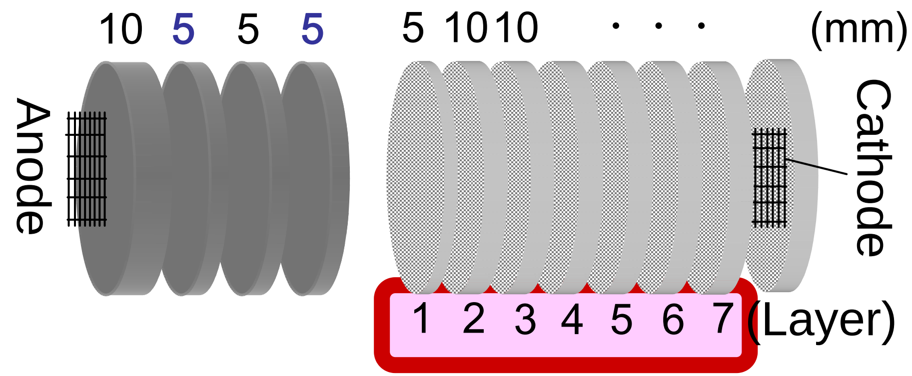
Reduction of leaching

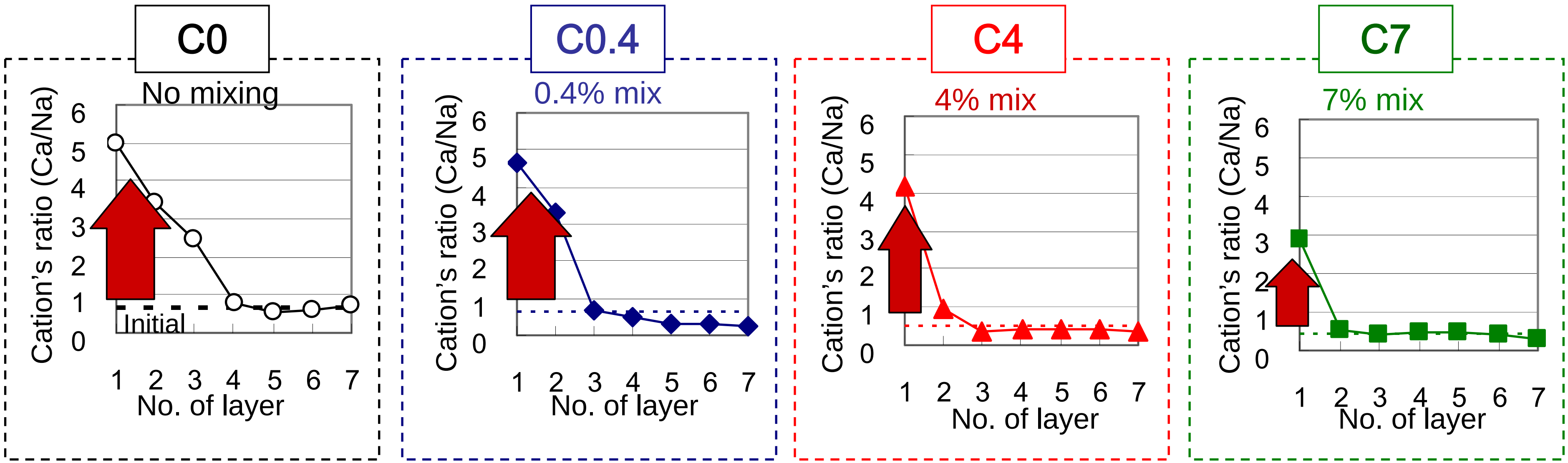


Result of Cations: Degradation of bentonite

Bentonite sand mixture

Swelling capacity
Cation concentration





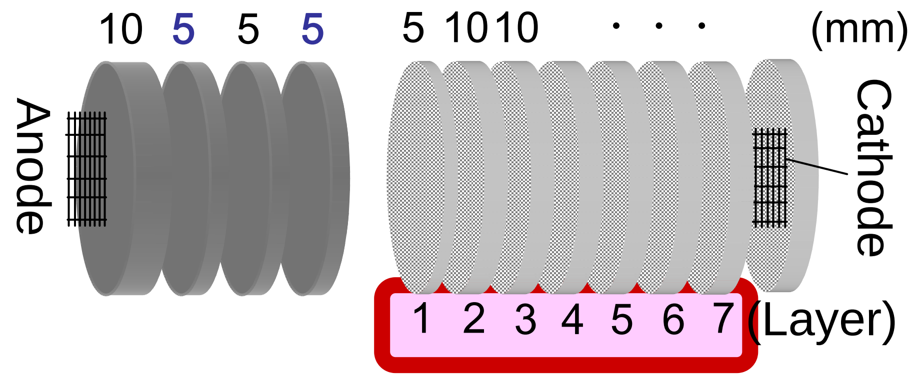
In specimen without NaHCO3, cation's ratio increased at the surface layers

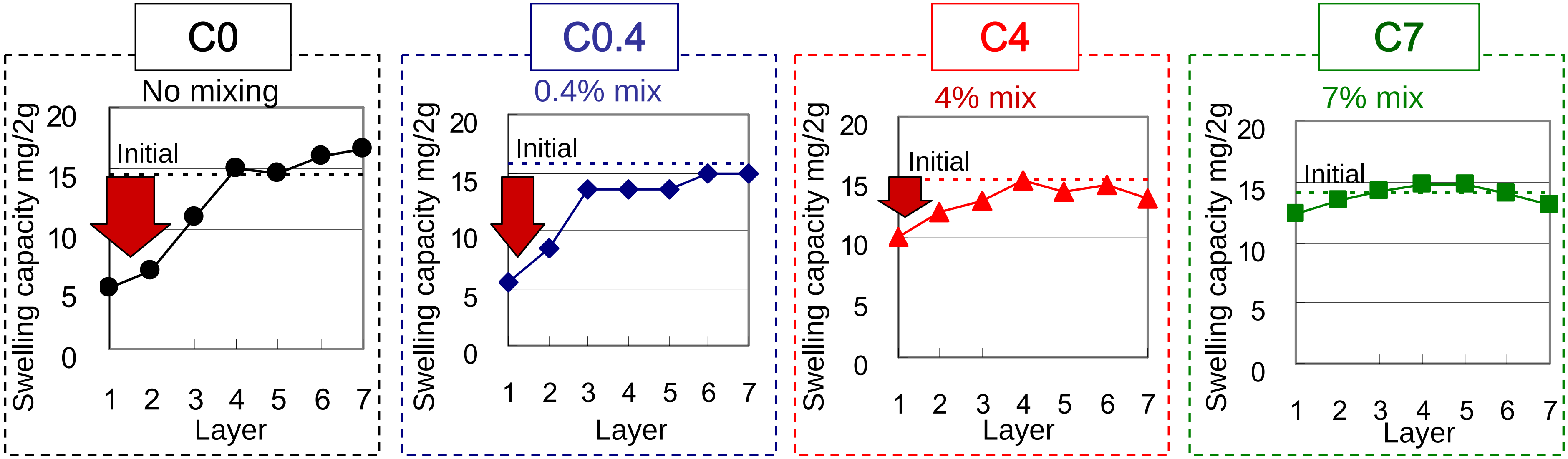
In specimens with large $NaHCO_3$, **increase in cation's ratio were significantly reduced**

Result of swelling capacity: Degradation of bentonite

Bentonite sand mixture

Swelling capacity
Cation concentration





In specimen without $NaHCO_3$, swelling capacity decreased at the surface layers

In specimens with large $NaHCO_3$, decrease in swelling capacity were significantly reduced

Next topic

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4. **Conclusions**

Conclusions

In this study, the long-term durability of the engineered barriers system was investigated by the migration technique.

Firstly, the effect of dry density of bentonite-sand mixtures was investigated.
The experimental results showed the use of the bentonite sand mixture having high dry density was effective with regard to the reduction in the risk of the alteration.

Secondly, the effect of mixing of $NaHCO_3$ to the bentonite-sand mixture was investigated.
The experimental results showed the mixing of $NaHCO_3$ clearly reduced the degradation of cementitious materials and bentonite because of precipitation of $CaCO_3$.

Thank you for your kind attention!

Specimens

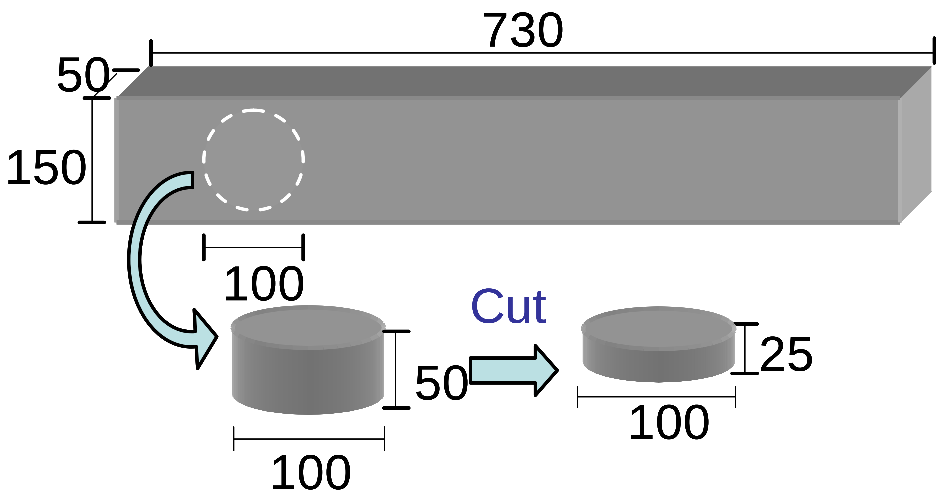
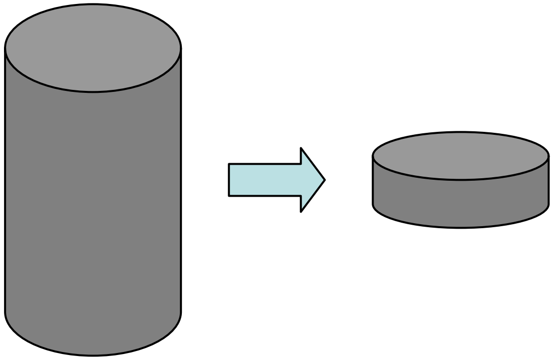
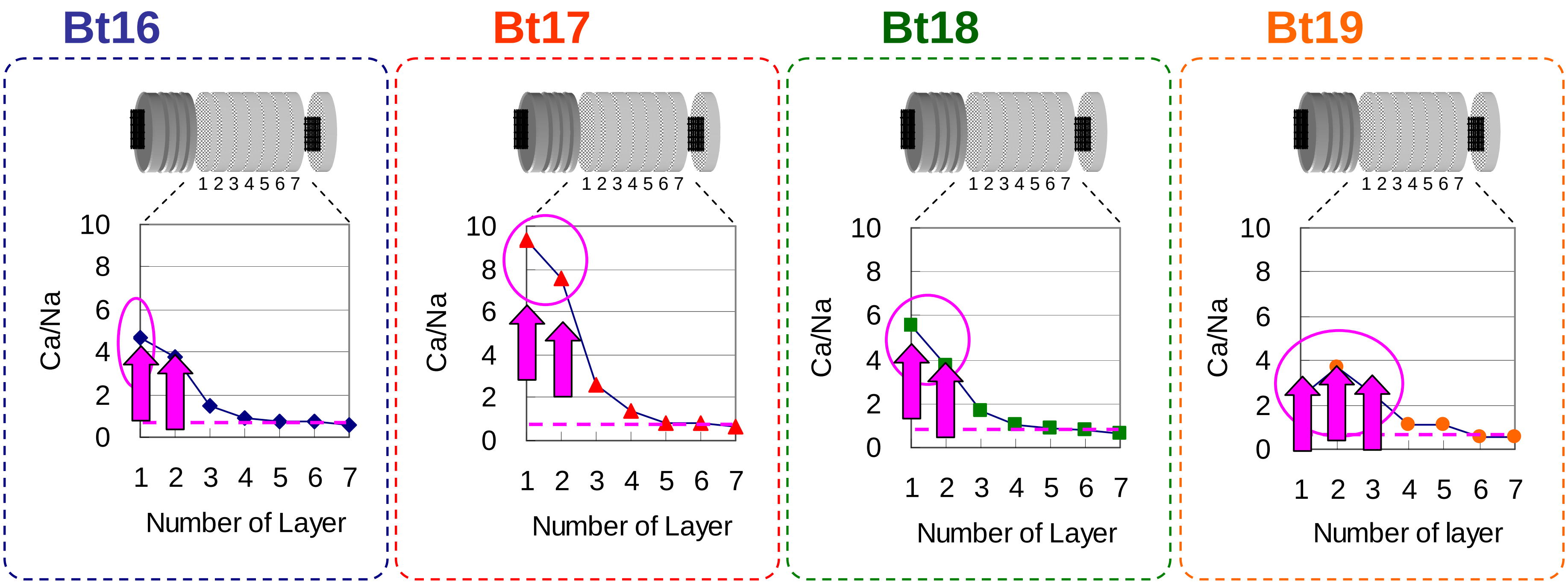


Fig. 1 Dimensions of concrete (mm)



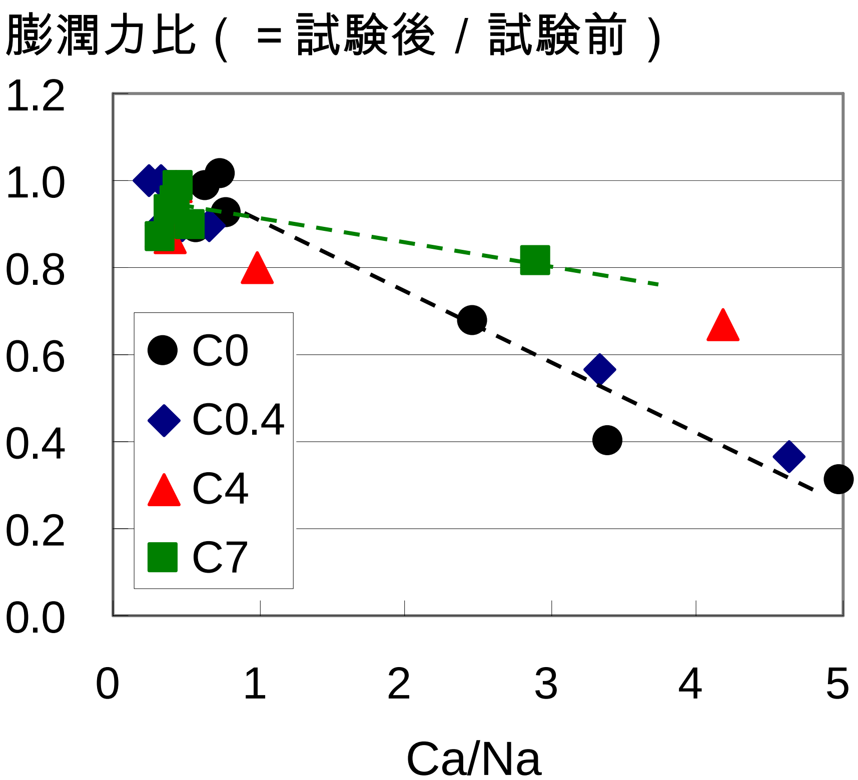
Result of cation's ration in bentonite

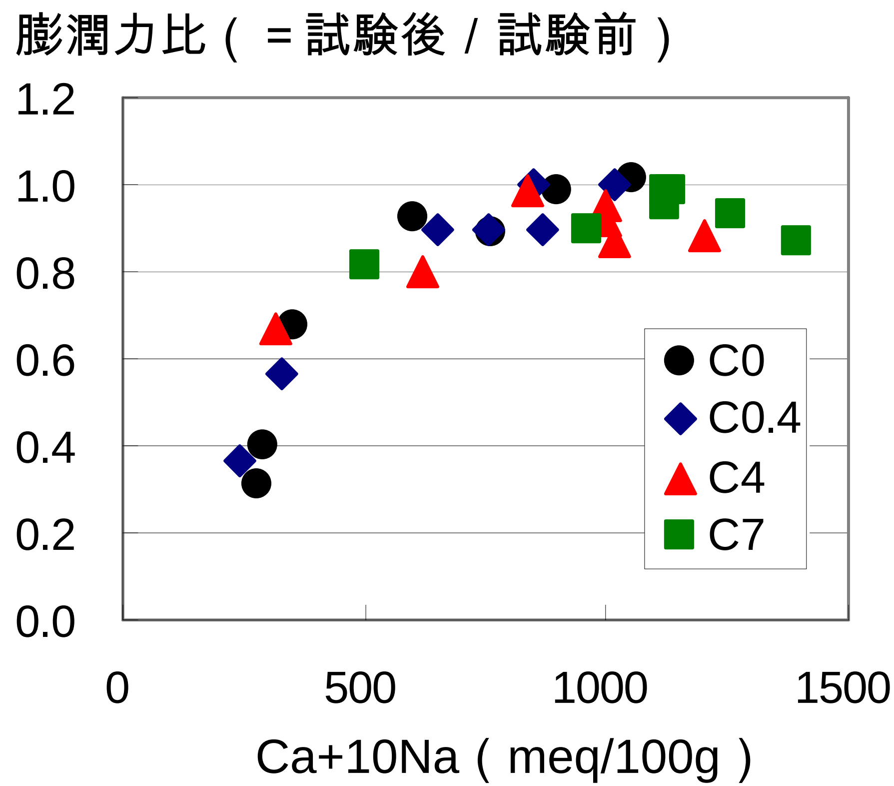


Concrete side:

Swelling capacity decrease with increase in Ca/Na → Change to Ca-type

ベントナイトのおける膨潤力と陽イオン量の関係

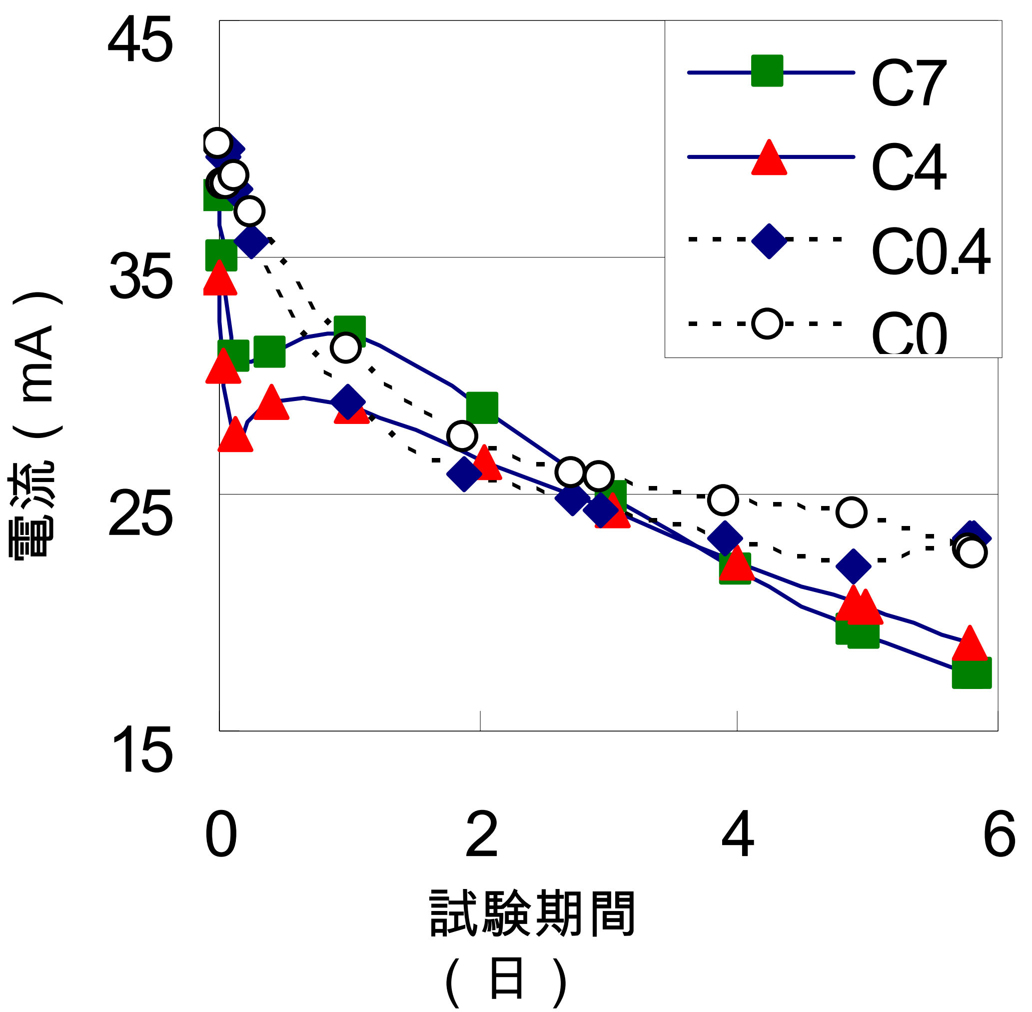


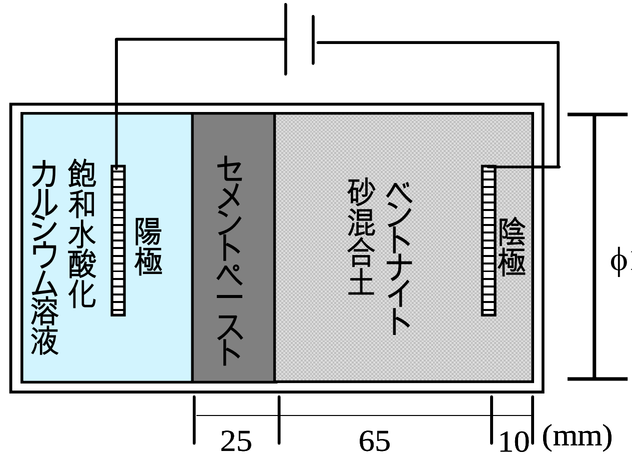


炭酸水素ナトリウム混合では
Ca/Naが高いが膨潤力低下はわずか

CaとNaの膨潤への貢献度合いを
足し合わせることにより膨潤力を
統一的に評価できる可能性も

電流の経時変化



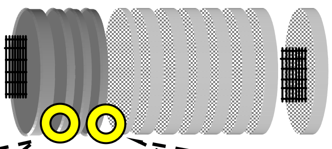


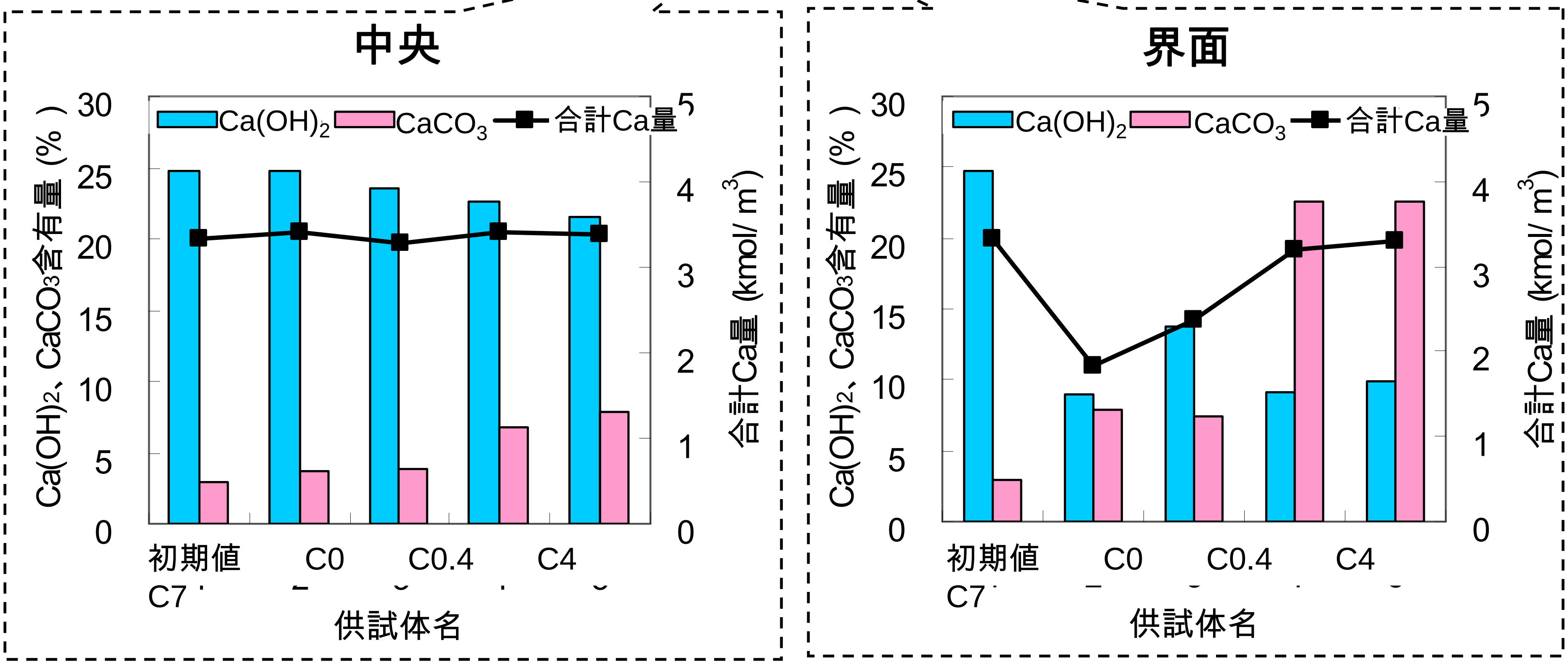
電気泳動試験結果

供試体名	試験期間 h	積算電気量 kC
C0	140	13.7
C0.4		13.0
C4		12.3
C7		12.7

合計Ca量の分布

合計Ca量：
セメントペースト単位体積あたりの$Ca(OH)_2$と$CaCO_3$の物質量の和

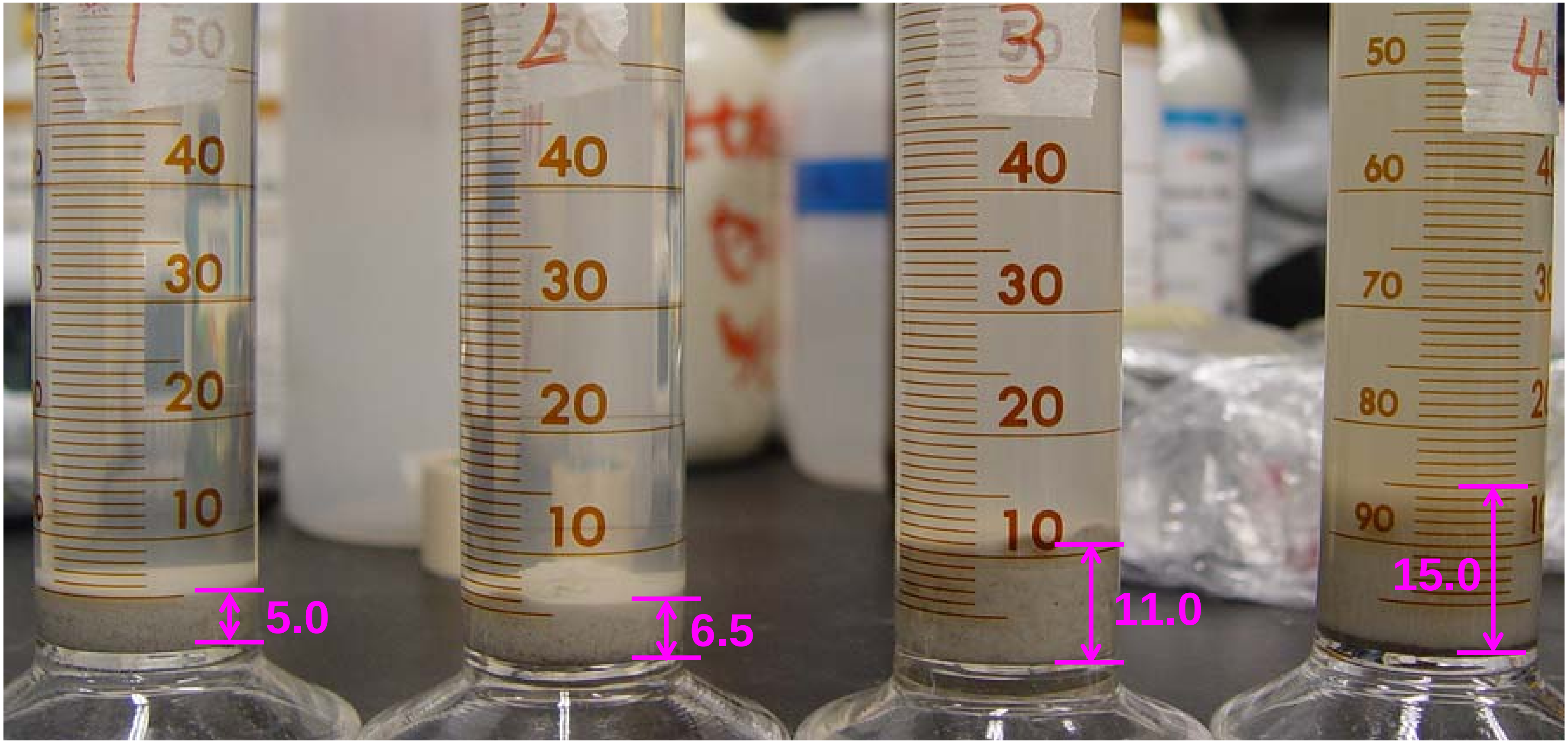




ベントナイトの試験

日本ベントナイト工業標準試験方法に準拠

浸出陽イオン量・・・ CaイオンとNaイオンの浸出陽イオン量比（Ca/Na）で評価
膨潤力・・・ 150μmに粉砕したベントナイト砂混合土の試料2.0gを，蒸留水100mlを入れた100mlのメスシリンダーに加え，24時間静置後，容器内に堆積した試料の見掛け容積を読みとることで測定



膨潤力測定例（C0の1～4層目）

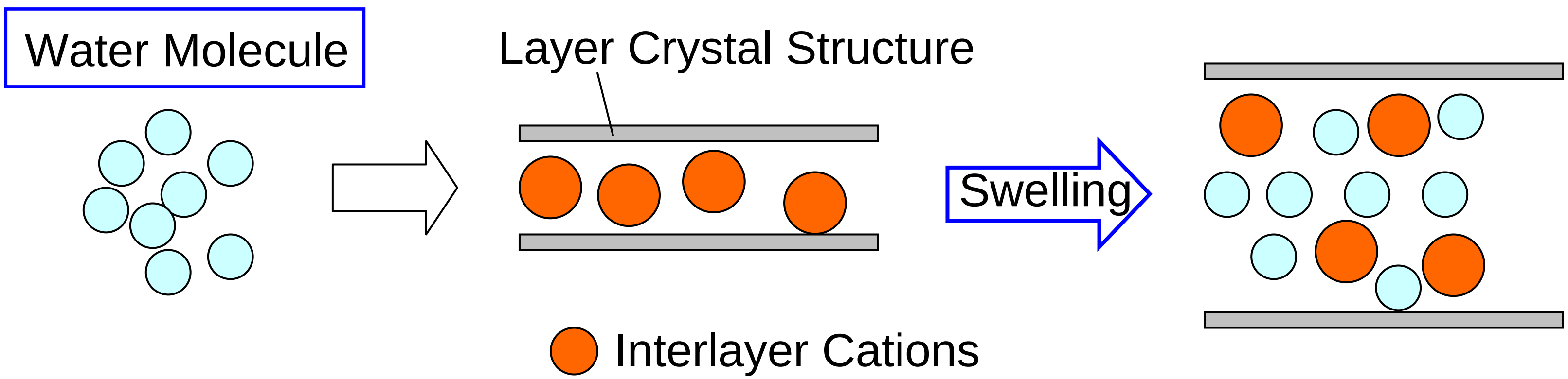
ベントナイトにおける浸出陽イオン量

表 - 6 浸出陽イオン量測定の結果

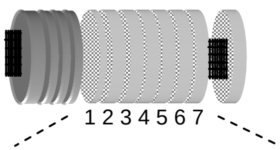
供試体名	イオン	浸出陽イオン量 meq/ 100g							
		初期値	電気泳動試験後						
			1層目	2層目	3層目	4層目	5層目	6層目	7層目
C0	Ca	40.5	92.3	73.3	69.9	43.8	41.0	53.0	73.0
	Na	59.3	18.6	21.5	28.4	55.5	72.0	84.3	97.8
C0.4	Ca	41.0	77.5	82.9	40.1	34.3	27.4	26.9	25.4
	Na	66.0	16.7	24.9	61.1	71.9	83.9	82.5	99.4
C4	Ca	42.9	92.8	55.1	37.6	35.1	40.3	39.8	38.9
	Na	93.4	22.2	56.2	98.1	80.4	95.6	95.7	116.4
C7	Ca	44.2	112.8	48.9	46.4	48.3	48.0	46.8	43.6
	Na	113.4	38.8	91.2	107.3	108.3	107.4	121.0	135.1

Swelling Capacity

Main component is montmorillonite

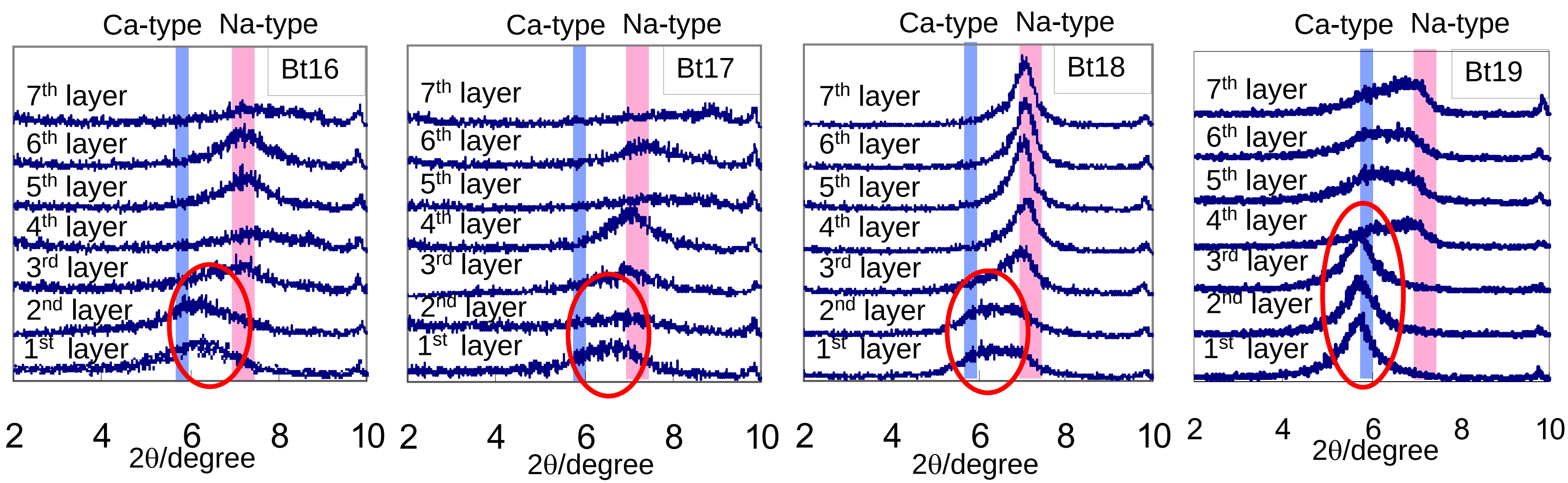


Results of XRD (Bentonite)



Peak range of angle of reflection in XRD curves shows type of bentonite (Kurosawa, 2002)

Na-type $2\theta=7.0\sim7.5°$	Ca-type $2\theta=5.7\sim6.0°$



Bt19: 1st to 3rd Layer → **Ca-type**

Bt16, **Bt17**, **Bt18**: 1st and 2nd Layer → Process of change to **Ca-type** ?

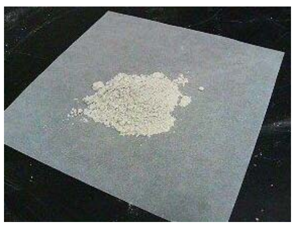
⇩

Changing of type start from concrete side

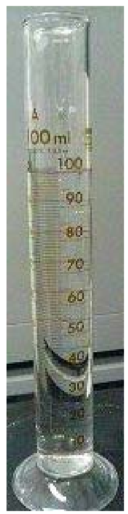
Cations and Swelling Capacities (Bentonite)

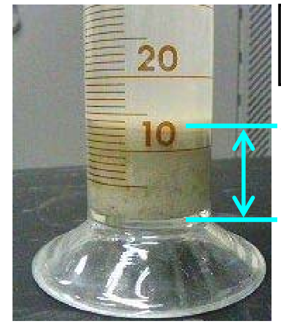
Method of Measurement of Swelling capacity

2g add to 100ml water



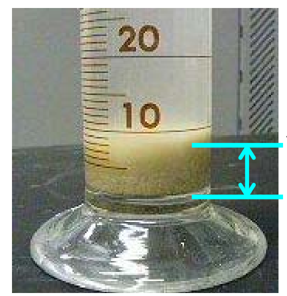
Bentonite Sand
Mixture Powder
(150μ)





Na-type

Swelling capacity
13 ml/2g



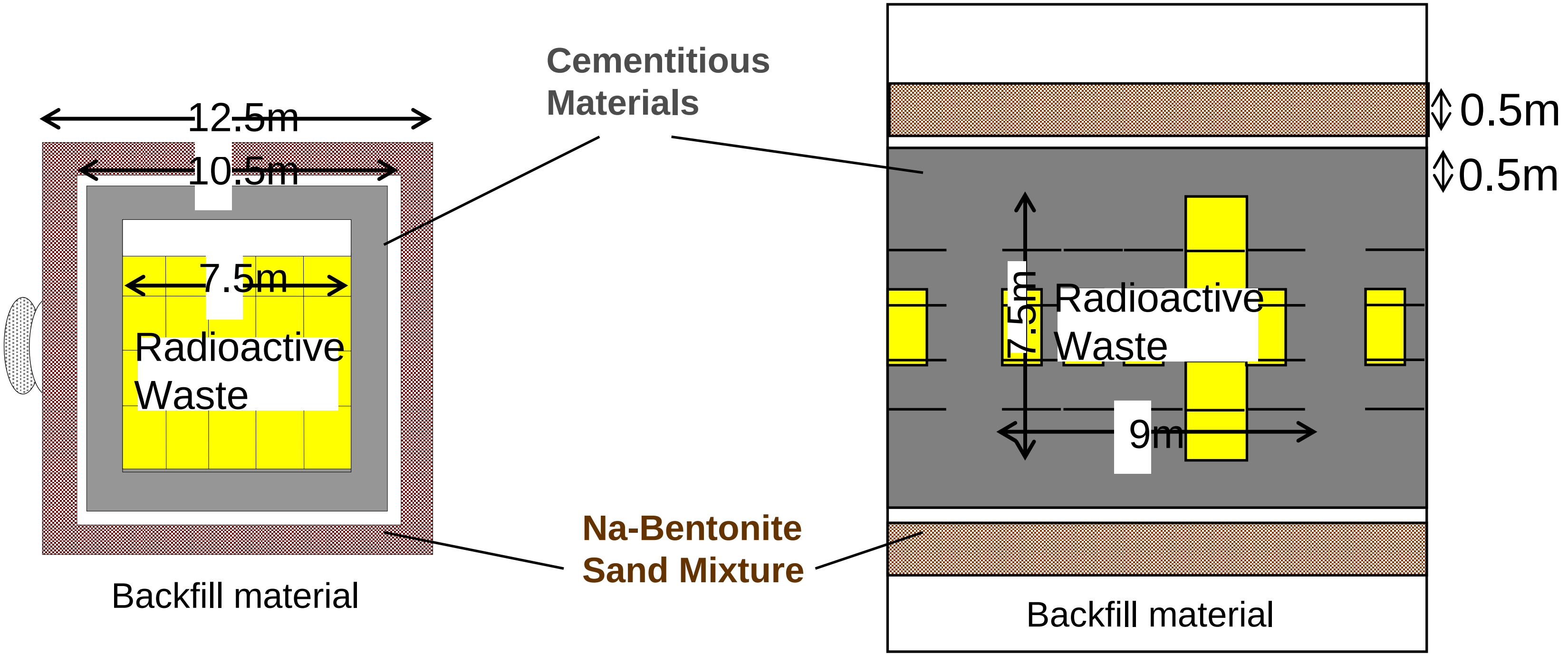
Ca-type

Swelling capacity
8 ml/2g

After leaving for 24 hour

Swelling Capacity Ratio: Swelling capacity divided by mean value of swelling capacity of 6th and 7th in each layer

Size of Geological Disposal Structure (Low-Level)



A Vertical Section

A Horizontal Section

Size of a section is 7.5m wide, 7.5m high, and 9m deep.