CESIUM CHEMICAL DESORPTION FROM FUKUSHIMA VERMICULITE

T. KOBAYASHI, S. SUZUKI, T. YAITA
ACTINIDE COORDINATION CHEMISTRY
QUANTUM BEAM SCIENCE DIRECTORATE

OUT LINE

- Background of this work
- Cs chemical desorption(Cs-133/Cs-137)
  - vermiculite(Fukushima)
  - biotite(india)/montmorillonite(kunipia F)
- Strategy of Cs chemical desorption
  - Ion exchange
  - destruction of clay minerals
- Future work

Background ①

@ Decontamination

reduce radiation dose risk.

⇒ more than 300,000 peoples take refuge.




× 18

2013.7.30
by MOE

remove huge contaminated soils.

@ Volume reduction of contaminated soils

(Cs recover from contaminated soils)

- Understands Cs sorption/desorption with soils.
- Develops new effective desorption methods.

Background ②

@ radiocesium sorption with soils.

property of Fukushima soils :

mica(a,b), smectite(vermiculite, montmorillonite)(a,b), kaolinite(a), hornblende(a), chlorite(b), etc.

a. N. Kozai *et al.,* J. Nuc. Sci. Tech. **49**, 473 (2012).
b. H. Quin *et al.*, Geochemical Journal **46**, 297 (2012).

@ evaluation of Cs sorption/desorption with mica, smectite.

main clay minerals in Fukushima.

Cs strongly adsorbed depending on layer charge.

FES (flayed edge site) strongly adsorbed Cs

evaluate desorption with chemicals

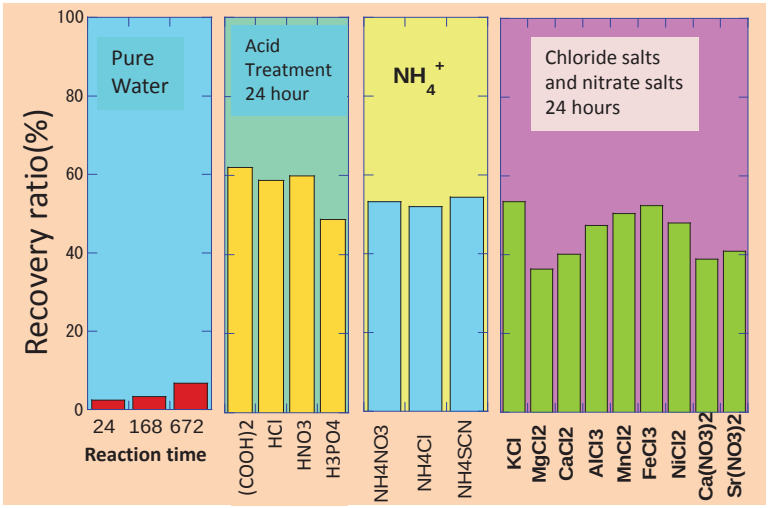
⇒ volume reduction

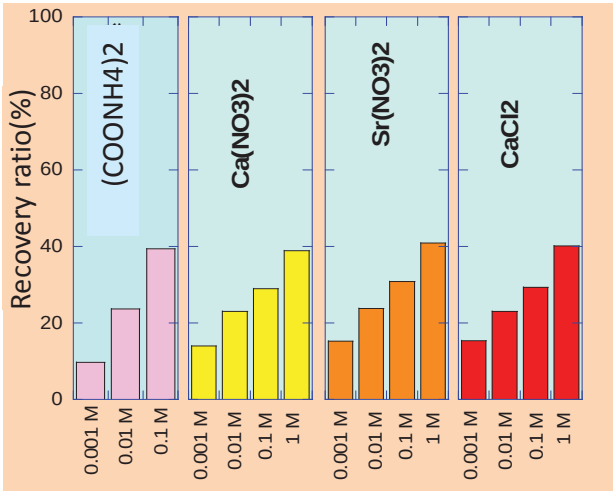
⇒ development of new methods

Cs desorption from Vermiculite ① (chemical reagent)

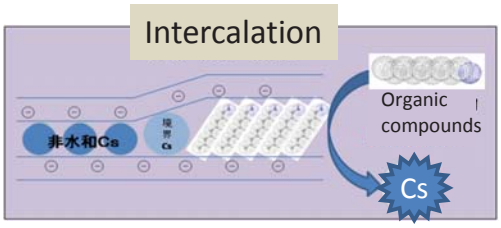
Cs maximum sorption with vermiculite :
(**4.9 mg** Cs/ 100 mg vermiculite)

Experimental
- ☛ volume ratio : 100 (Vermi;100 mg, solution:10 mL
- ☛ chemicals concentration :
- ☛ adsorption time :
- ☛ pretreatment : filtration(0.45 μm) / dilution(HNO_3)
- ☛ analytical method : ICP-MS (PerkinElmer, NexION 300D)

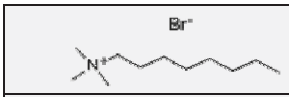
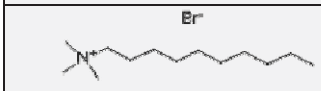
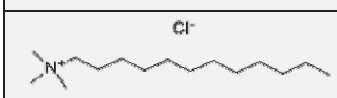
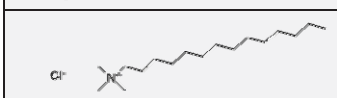
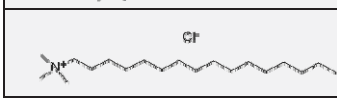


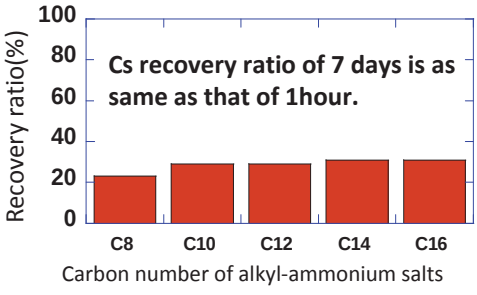


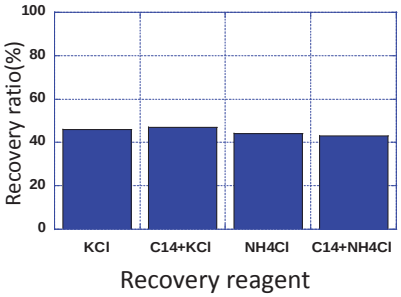
Cs desorption from Vermiculite ② (Intercalation)

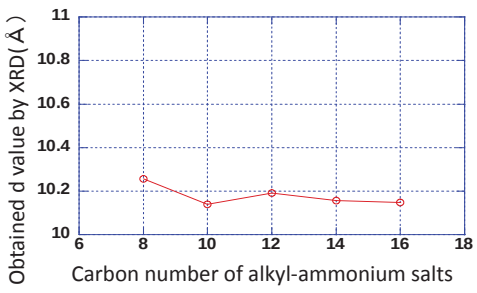


Experimental
- ☛ solid :liquid ratio : 200 mg : 20 mL
- ☛ concentration :
- ☛ temperature :
- ☛ reaction time :
- ☛ filtration : 0.45 μm
- ☛ analysis : ICP-MS
- ☛ KCl、NH4Cl conc. : 1.0 M

	Octyltrimethylammonium bromide **C8**
	Decyl trimethyl ammonium b romide **C10**
	Dodecyl trimethyl ammonium chloride **C12**
	Tetradecyl trimethyl ammonium chloride **C14**
	Hexadecyltrimethylammonium chloride **C16**

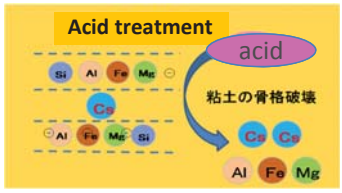


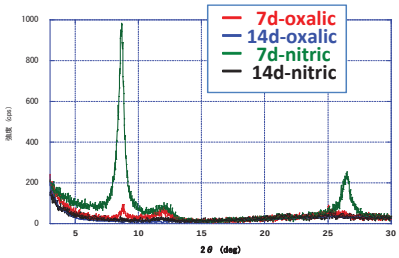


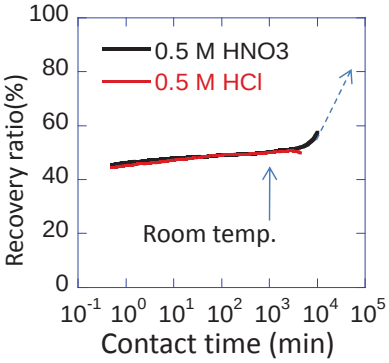


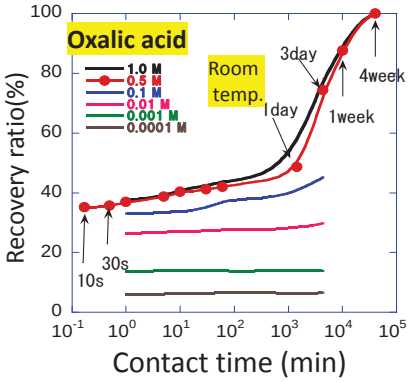
Cs desorption from Vermiculite ③ (acid + 95℃)

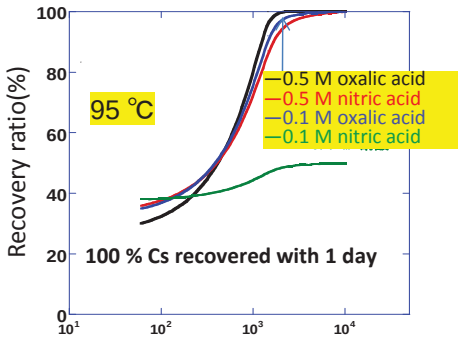
Experimntal
- Vermiculite : soln : 100 mg : 10 mL
- Cs concentration :
- adsorption time :
- pre-treatment : 0.45μm filteration and dilution
- analysis : ICP-MS











100 % Cs recovery by room temp/1 month or 95℃/1day.

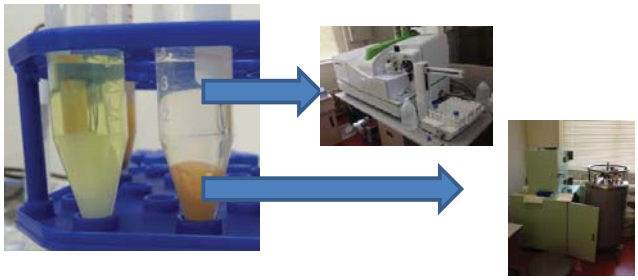
	Si	Al	Fe	Ca	K	Mg
Vermiculite	27.2	14.9	37.3	1.30	8.44	6.66
1M Oxalic acid 1day	40.5	17.4	25.6	0.219	7.34	4.45
1M Oxalic acid 7 days	74.6	15.2	7.14	0.224	0.69	0.37
1M NH4Cl 1week	27.3	15.1	37.9	-	6.9	5.28
0.1 M C14 1 M NH4Cl	23.5	12.5	43.7	0.23	6.52	4.49

	1 M Oxalic acid(1day)	1 M oxalic acid (7day)
Fe/Si	0.68 → 0.32	0.68 → 0.05
Al/Si	0.57 → 0.41	0.57 → 0.21
Mg/Si	0.28 → 0.13	0.28 → < 0.01
K/Si	0.22 → 0.13	0.22 → < 0.01

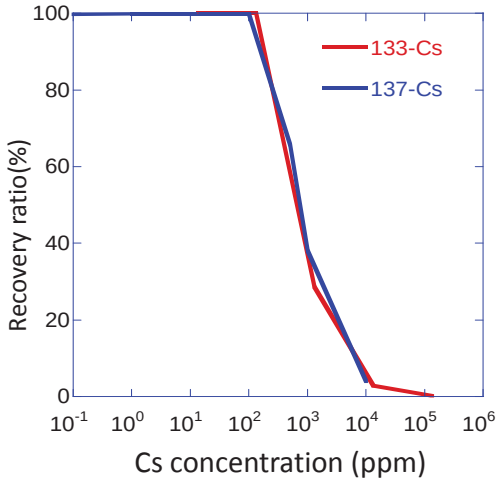
Cs-137/Cs-133 sorption and desorption with Vermiculite

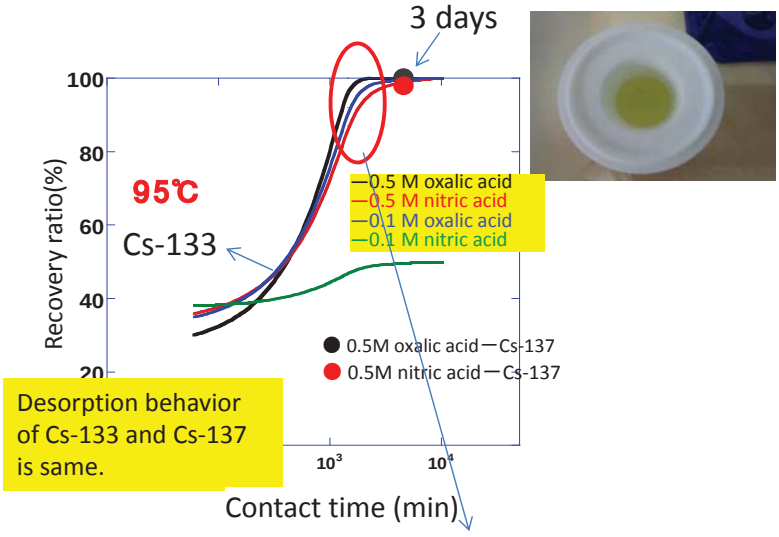
pH dependency
- solid : solution = 50 mg : 5 mL
- pH : value = 1～12
- activity :
- contacting time :
- Pre-treatment : 0.45 μm Filter
- analysis : Ge semiconductor detector, ICP-MS

Cesium specific activity
Cs-137 : 3.2x10^{12} Bq/g



Cs-137 solution preparation
Spiked Cs-137 in each concentrated Cs-133 solution
And leaved a sample for one week.





Desorption behavior of Cs-133 and Cs-137 is same.

The oxalic acid is enough in a day to recover 100% of Cs.

Summary (Cs desorption from 3 kinds of clay minerals)

sample	Acid treatment (95℃・1W)	Ammonium salts	Alkaline /alkali earth group	Intercalation	Layer change with intercalation
Vermiculite (Fukushima)	～60% (100%)	～57%	～55%	～30%	×
Biotite (India)	～22% (23% ?)	～18%	～18%	～12%	×
Montmorillonite (Kunipia F)	～38% (60%)	～42%	～40%	～55%	The infinite expansion?

Vermiculite : 100% Cs recovered easily with 95 ℃/ acidic condition.
Biotite : about 20 % Cs recovered with several condition. In case of 95%/acidic condition, Fe-oxalic complex exists solid sample. This Fe-oxalic complex might adsorb Cs.
Montmorillonite : Interlayer of montmorillonite expanded infinitely using alkyl ammonium salts. However, Cs recovery ratio is about 60 %.

JAEA Tokai Research Center

Thank you for your attention!!

ご清聴ありがとうございました。

SPring-8