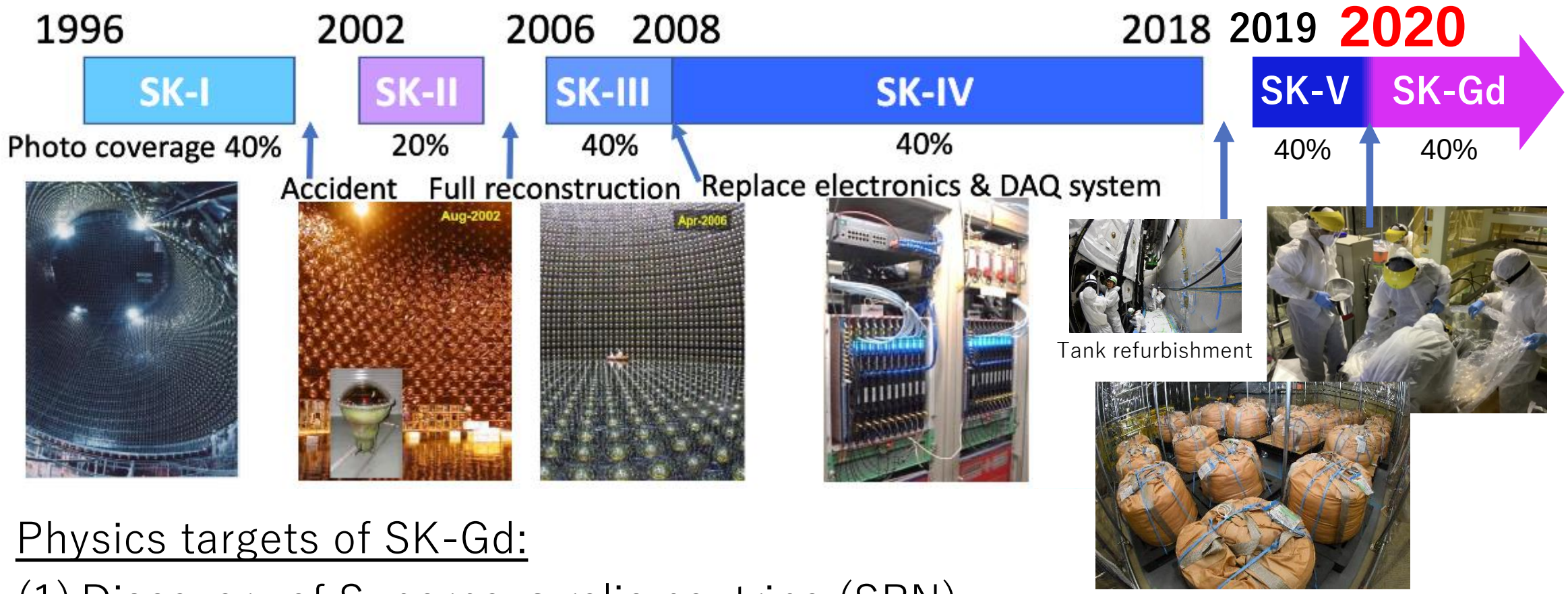
令和3年度　共同利用発表会

ICP質量分析器を用いた，スーパーカミオカンデ等，地下実験のための極微量放射性不純物元素の測定

- 研究組織：
  - 岸本康宏（代表），市村晃一、後藤杏奈、古田雄貴（東北大）
  - **池田一得（東大神岡）（発表者）**
  - 坂口綾、高久雄一（筑波大）

- 採択額：
  - 物件費　88,000円，旅費 300,000円，総額388,000円
    - 研究支援ありがとうございました.

Super-Kamiokande Gd project just started



Physics targets of SK-Gd:

(1) Discovery of Supernova relic neutrino (SRN)

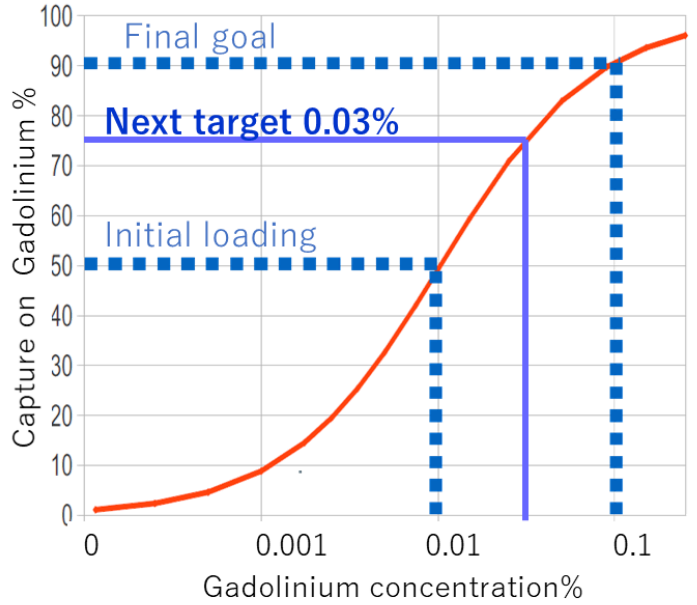
(2) Galactic supernovae (pointing accuracy, and Si-burning ν)

(3) Reduction of BG for proton decay, solar ν , or reactor ν

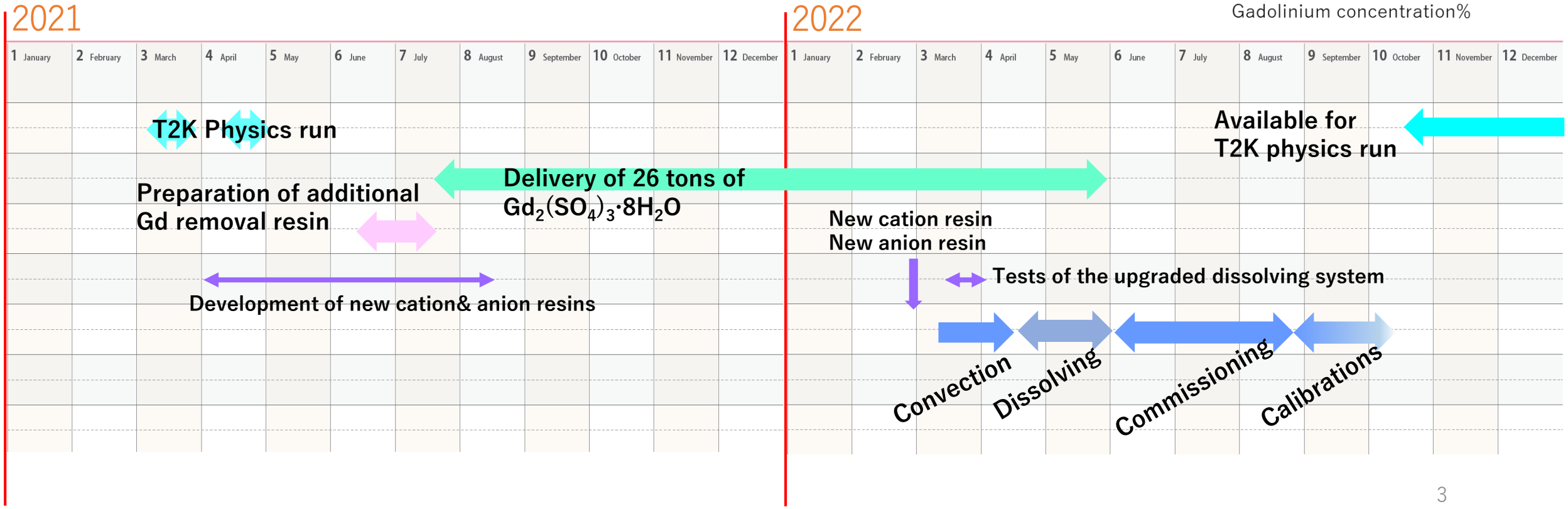
(4) Neutrino/anti-neutrino discrimination

Plans of SK-Gd

- Aiming to dissolve up to ~26 tons of additional $Gd_2(SO_4)_3 \cdot 8H_2O$ in 2022
  - Target Gd concentration: 0.03% (Currently 0.01%)
  - Gd capture efficiency: 75% (Currently 50%)



Current plan for the next Gd-loading



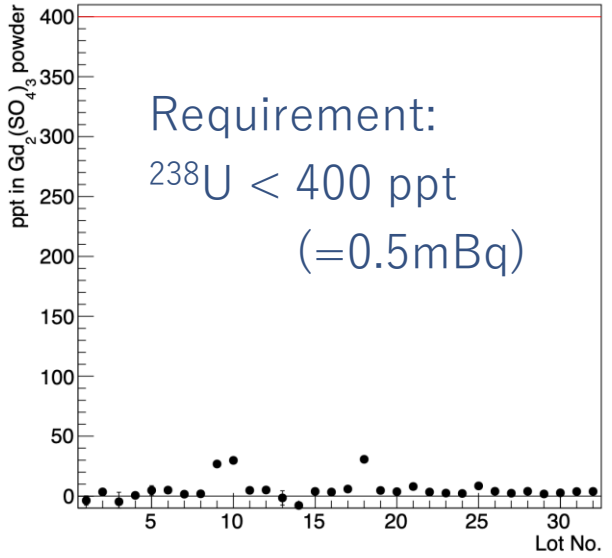
Toward 26 tons of clean $Gd_2(SO_4)_3 \cdot 8H_2O$

- Gd powder for the first loading was screened lot by lot at multiple sites:
  - ICP-MS: Kamioka
  - HPGe: Canfranc, Boulby and Kamioka
- ^{238}U, ^{232}Th, ^{235}U < requirements.
- ^{228}Ra was found to be ~1mBq/kg in latter half of the production.
  - Gd oxide which is the feedstock of Gd sulfate had more ^{228}Ra for such lots
  - Cannot find cleaner feedstock.

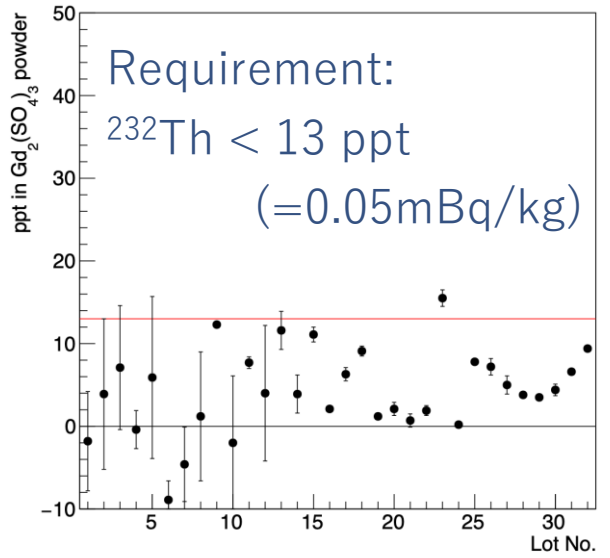
Radioactive impurities for $Gd_2(SO_4)_3$ powder [mBq/kg]

Chain	Isotope	SK-Gd requirements	
		For solar	For DSNB
^{238}U	^{238}U	—	< 5
	^{226}Ra	< 0.5	—
^{232}Th	^{228}Ra	< 0.05	—
	^{228}Th	< 0.05	—
^{235}U	^{235}U	< 30	—
	$^{227}Ac/^{227}Th$	< 30	—

U contamination (ICP-MS)



Th contamination (ICP-MS)



Status of making Gd for the next loading

- We found additional purification processes can reach the same level of RI as that of the first half of 13 ton (ex. Lot # 190302).
- Production of 26 tons of Gd sulfate has been started since June, 2021
  - Production rate : 2.5-3 ton per month
  - 26 tons will be ready by the end of May, 2022.

	U-238 early	U-238 late	Th-chain Ac-228 (=Ra-228)	Th-chain late	U-235 early	U-235 late
210301 New $Gd_2(SO_4)_3$	<2.8	<0.36	<0.28	<0.10	<1.44	<1.88
190302 For first loading	<9.8	<0.32	<0.35	<0.29	< 0.42	< 0.92

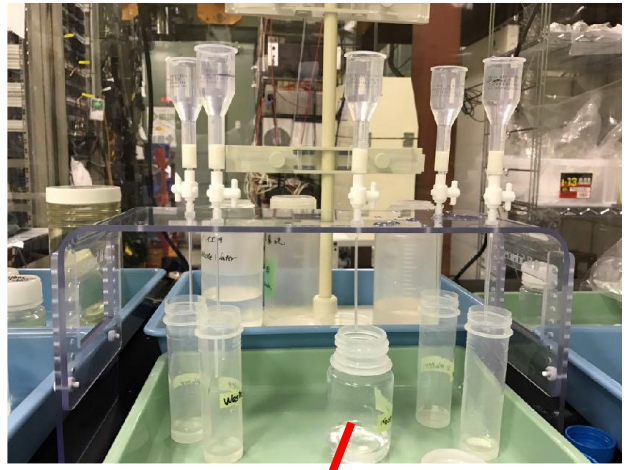
Evaluation of super-low level U/Th

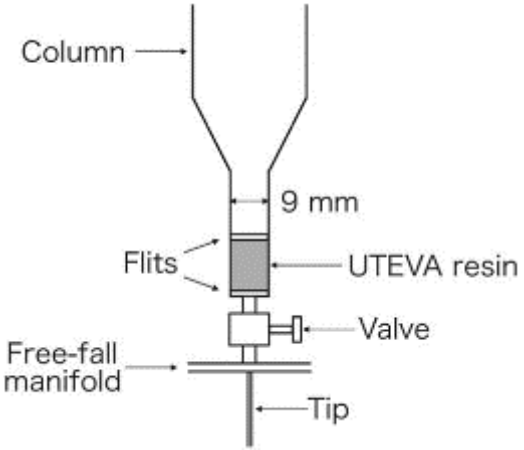
- We have developped a method to measure super-low level U/Th in Gd powder
- Requiements:

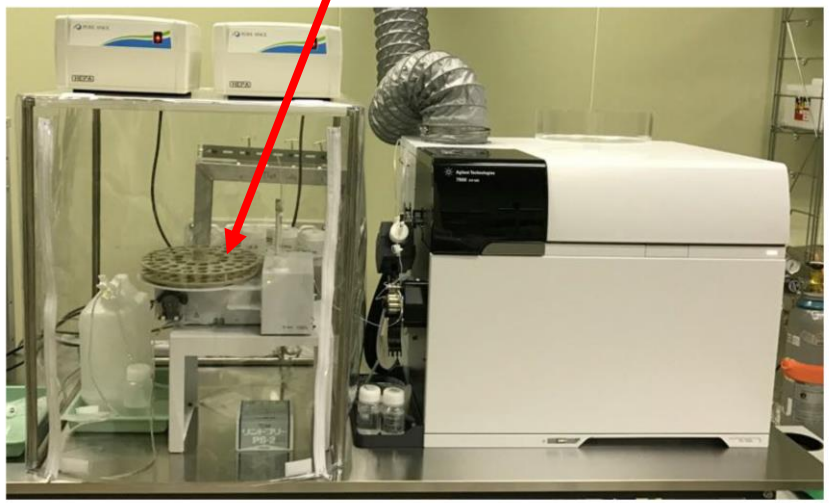
  ^{238}U < 400ppb (5mBq/kg),

  ^{232}Th< 12ppt (0.05mBq/kg)

  - Separation and extraction of U/Th from Gd solution using resin
    - To remove matrix effect of Gd
    - S.Ito et al. PTEP 2017 113H01







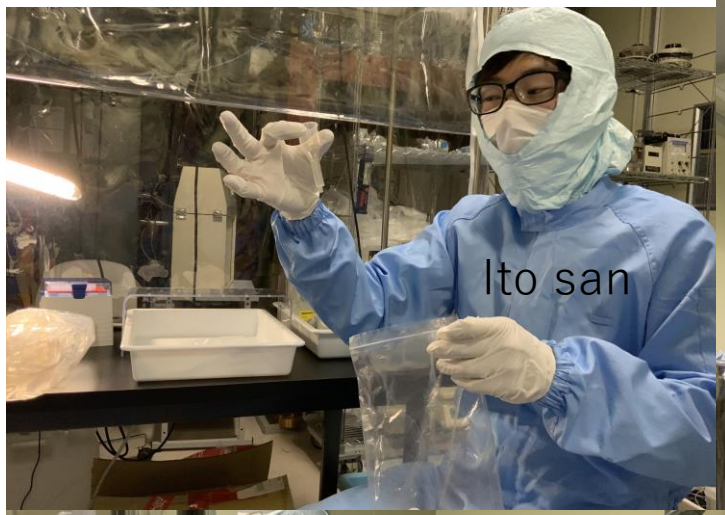
Auto-sampler is covered by clean booth.→**Class 100**

Education to young members

Expert (S.Ito san: Okayama->KEK) gave lectures for young people, so that more people can participate in the screening campaign.

Now two new members (+Ikeda) are working on the U/Th separation and ICP-MS measurements.

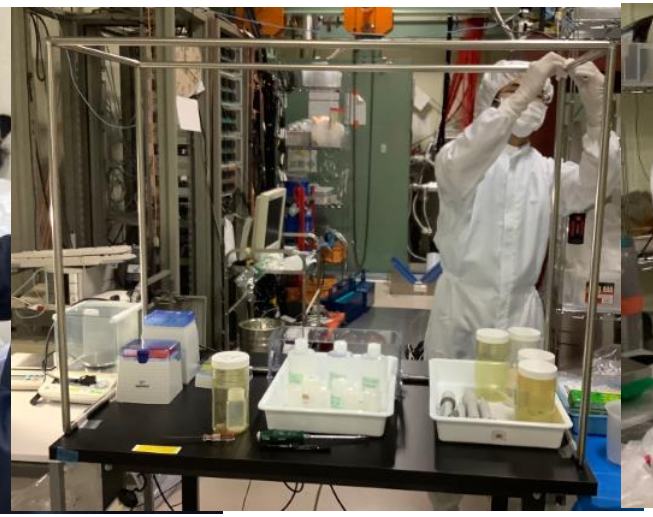
Need to catch up with Ito san's quality!

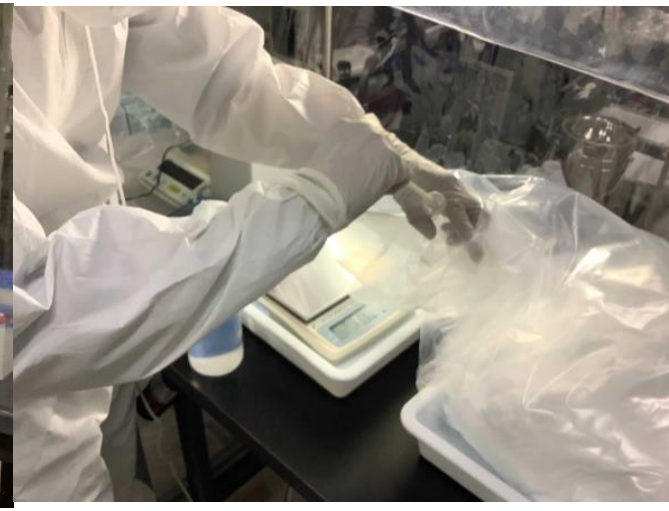


Lesson learned

↓Test with known clean Gd (sub ppt)

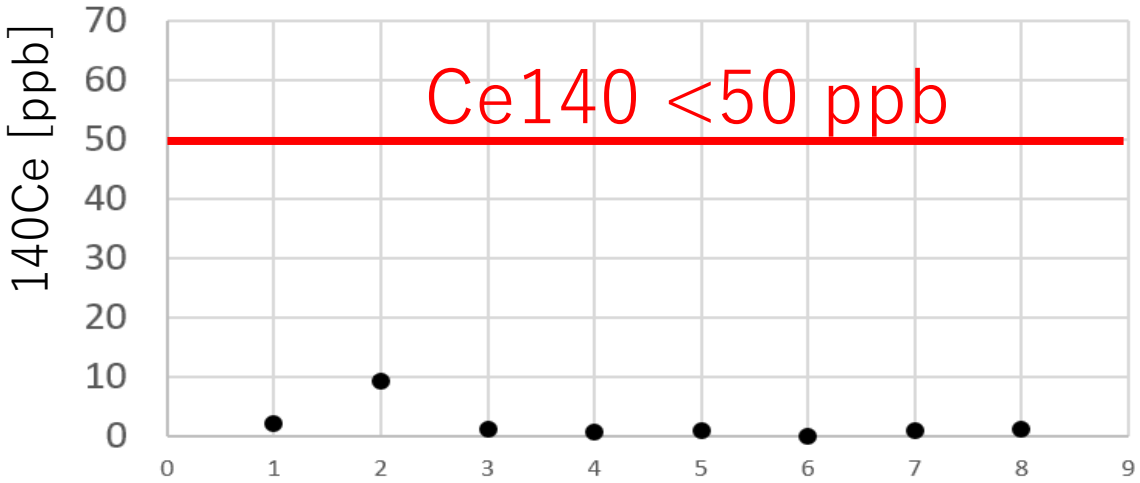
Date	Blank [ppt]	238Th[ppt]	Comments
2021/6/8	120	37.5	Resin wash was not perfect
2021/6/18	0.8	19.8	Procedure was correct
2021/7/20	1	18.5	All bottles were washed with correct procedure
2021/9/6	1	17	ICP-MS maintenance (annual) was done. Cleaned and tided up the room. HEPA filter for the clean booth Replaced the clean booth sheets.
2021/9/29	1	2.5	Sample powder from 10kg bag in the clean booth.
2021/9/29	0.5	1.6	Gd sulfate concentration of the prodedure 2%->4%

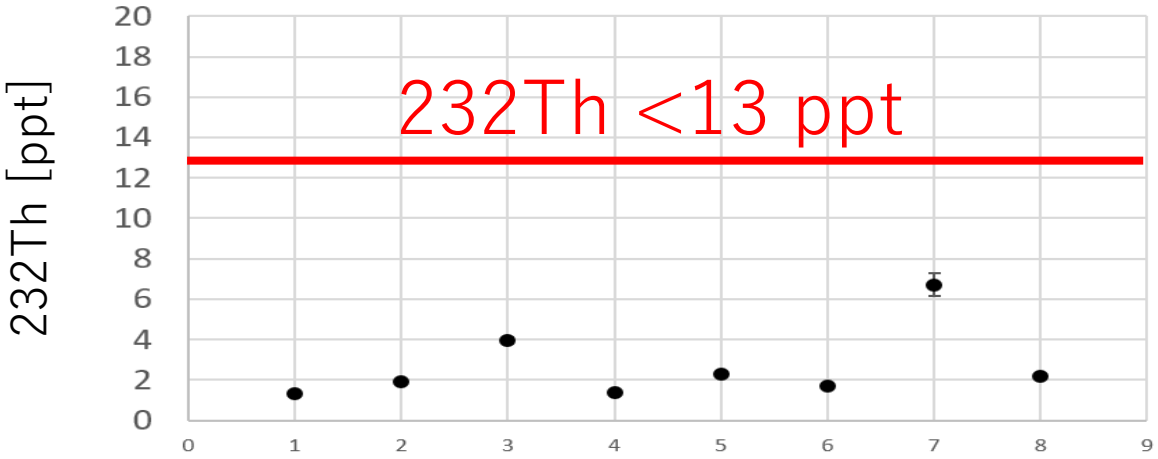



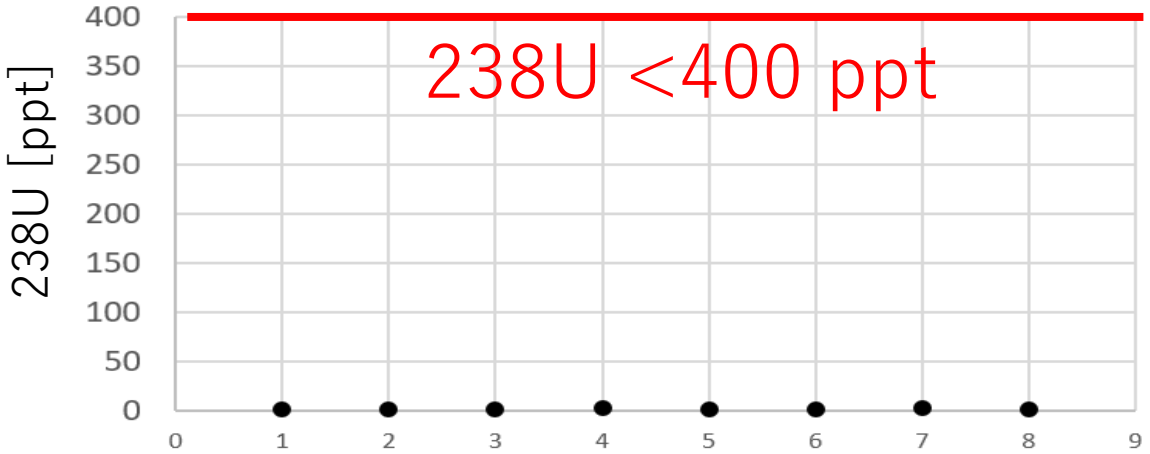


Status of the production and the screening

- ~1/3 of production finished.
- Check with ICP-MS is on going.
- Confirmed that received samples meet our requirements
- Production will finish by the end of May 2022.







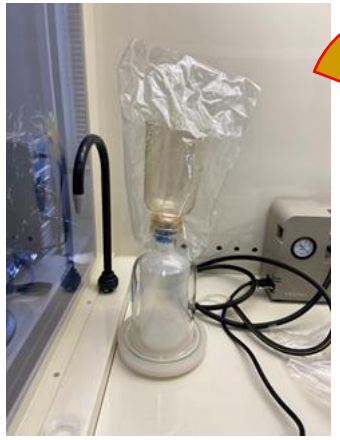
極微量放射性核種 測定法開発のための設備や実験（筑波大学）

Slide from Sakaguchi san

SK-Gdに使用する硫酸ガドリニウム中の極微量放射性核種分析

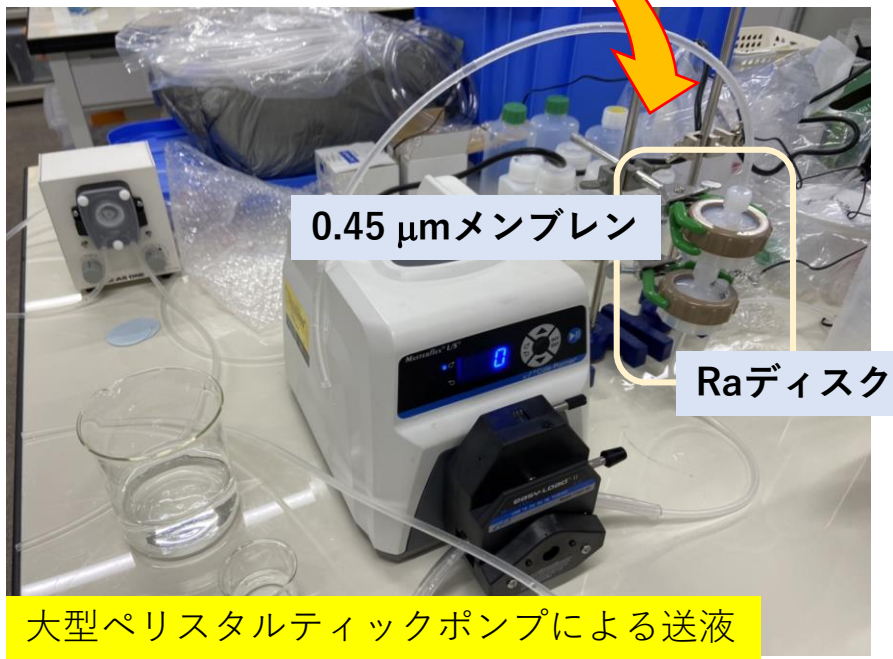
ICP-QQQ-MS/Ge半導体検出器による Ra-226測定のための方法を検討

現行の濾過装置



Raディスクに硫酸Gd溶液を通すろ過装置

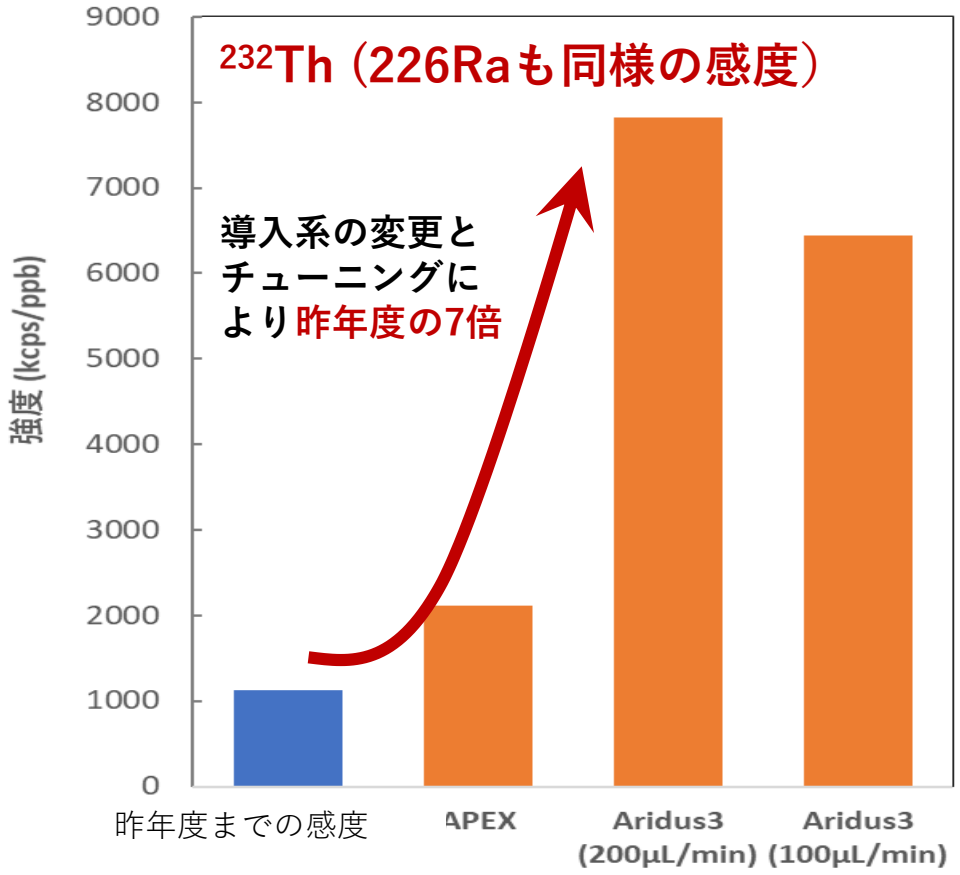
溶液を移し替える手間を省き、安全に簡単に吸着



大型ペリスタルティックポンプによる送液

Raディスクへの吸着方法の簡便化

ICP-QQQ-MSの重元素高感度



Ra-226測定では...

必要な硫酸ガドリニウム溶液量が約 **9 L**まで減少

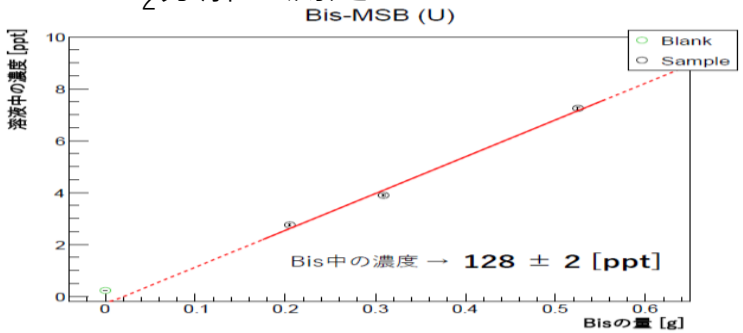
極微量放射性核種 測定法開発のための設備や実験（筑波大学）

Slide from Sakaguchi san

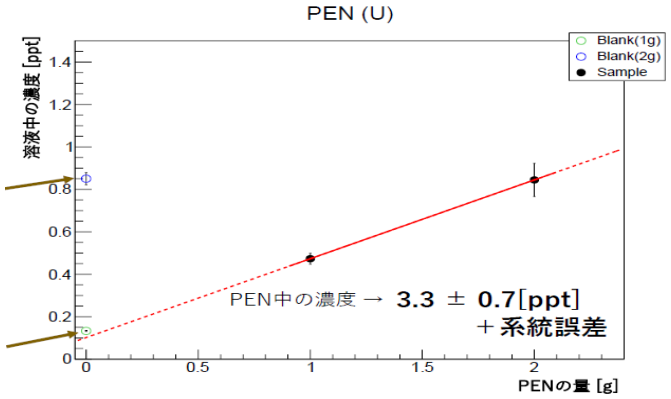
KamLANDで使用する樹脂中の極微量U, Th（＆安定元素不純物）分析
CANDLESで使用するCaF$_2$中のU, Th測定

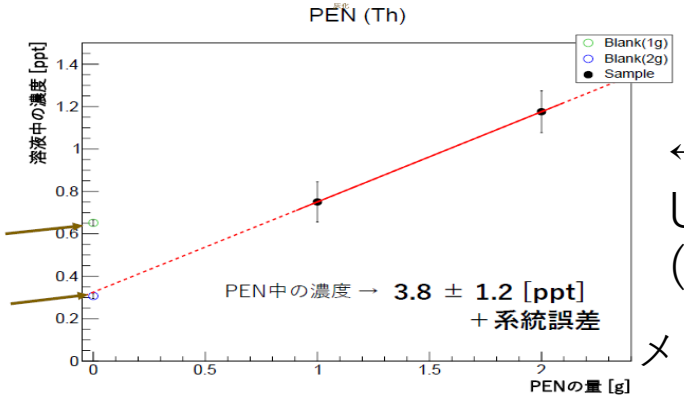
地下宇宙プロジェクト用ラボ!?

CaF$_2$分解＆測定



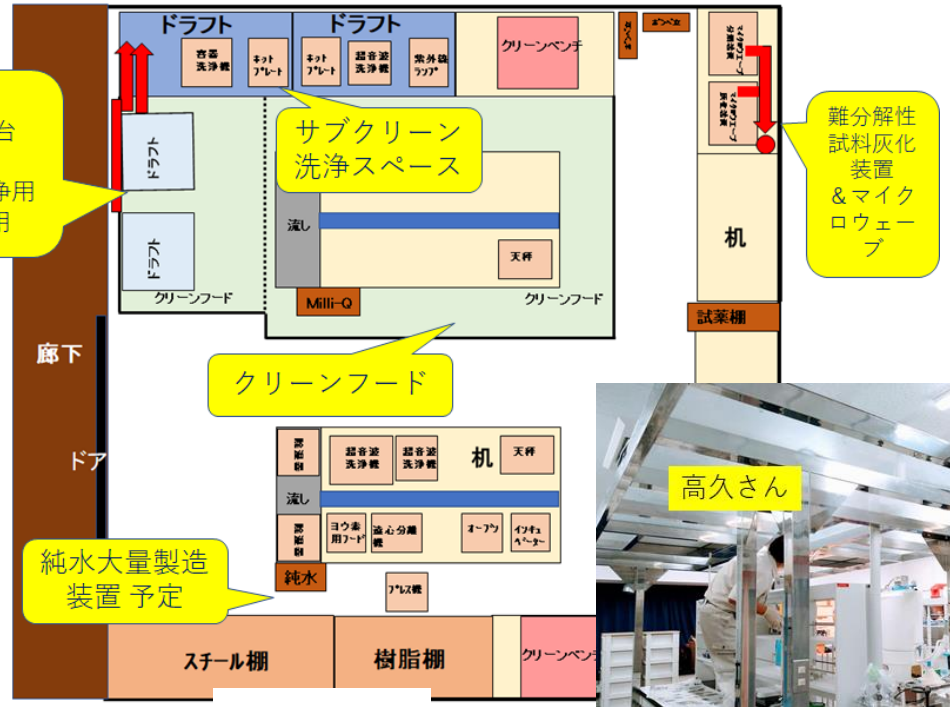
↑Bis-MSB中のU濃度



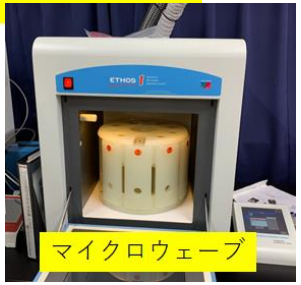


←試料処理後のPEN中
U, Th濃度測定結果
(ICP-QQQ-MS)

メインワーク(SK-Gd)より進んでいる疑惑



倉澤さん（東北大M1)を見守る
高久さん



Summary

- SK-Gd will be upgraded with another 26 tos of Gd-sulfate.
  - 26 tons of Gd sulfate powder will dissolved to SK water after GW this year.
    = ~ 0.03% Gd concentration
    = Gd capture efficiency: 75% (Currently 50%)
- We will finish the screening of Gd sulfate powder before that
  - ~ 1/3 of screening has finished so far
- Tsukuba team is also very active for SK-Gd and other underground related experiments.

RI in Gd powder before R&D

Requirement for each isotope assuming 0.2% Gd sulfate loading

Isotope	SRN	Solar	Before 2015
^{238}U	< 5	-	50
^{226}Ra	-	< 0.5	5
^{232}Th	-	< 0.05	
^{228}Ra	-	< 0.05	10
^{228}Th	-	< 0.05	100
^{235}U	-	< 3	32
$^{227}Ac/Th$	-	< 3	300

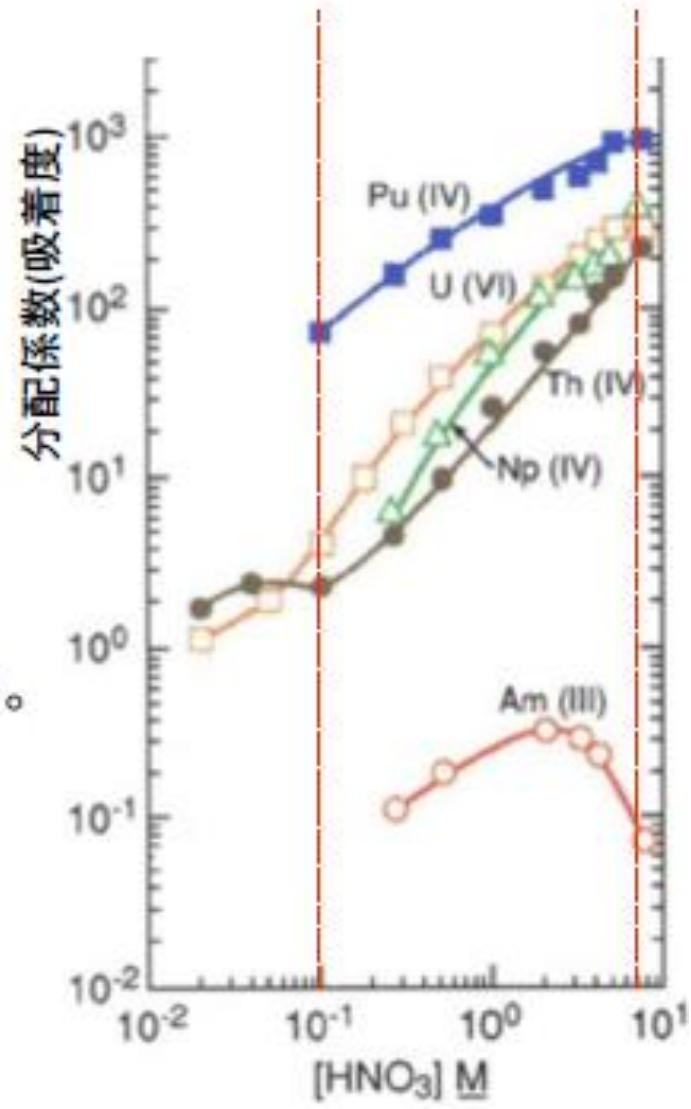
Unit：mBq/kg($Gd_2(SO_4)_3$) $8H_2O$

1/10 ~ 1/1000 reductions were needed!

U,Th 吸着樹脂

- 硫酸ガドリニウム中の微量のトリウム、ウランを化学分離して分析

- UTEVA resin
  - 6価のUや4価のアクチノイド(Th)は吸着。
  - 硝酸の濃度により吸着率が変わる。
    - → 7M 硝酸で吸着、0.1M 硝酸で溶離。
    - → Gdは3価の希土類なので、ひっつかない。



PTEP 2017 113H01

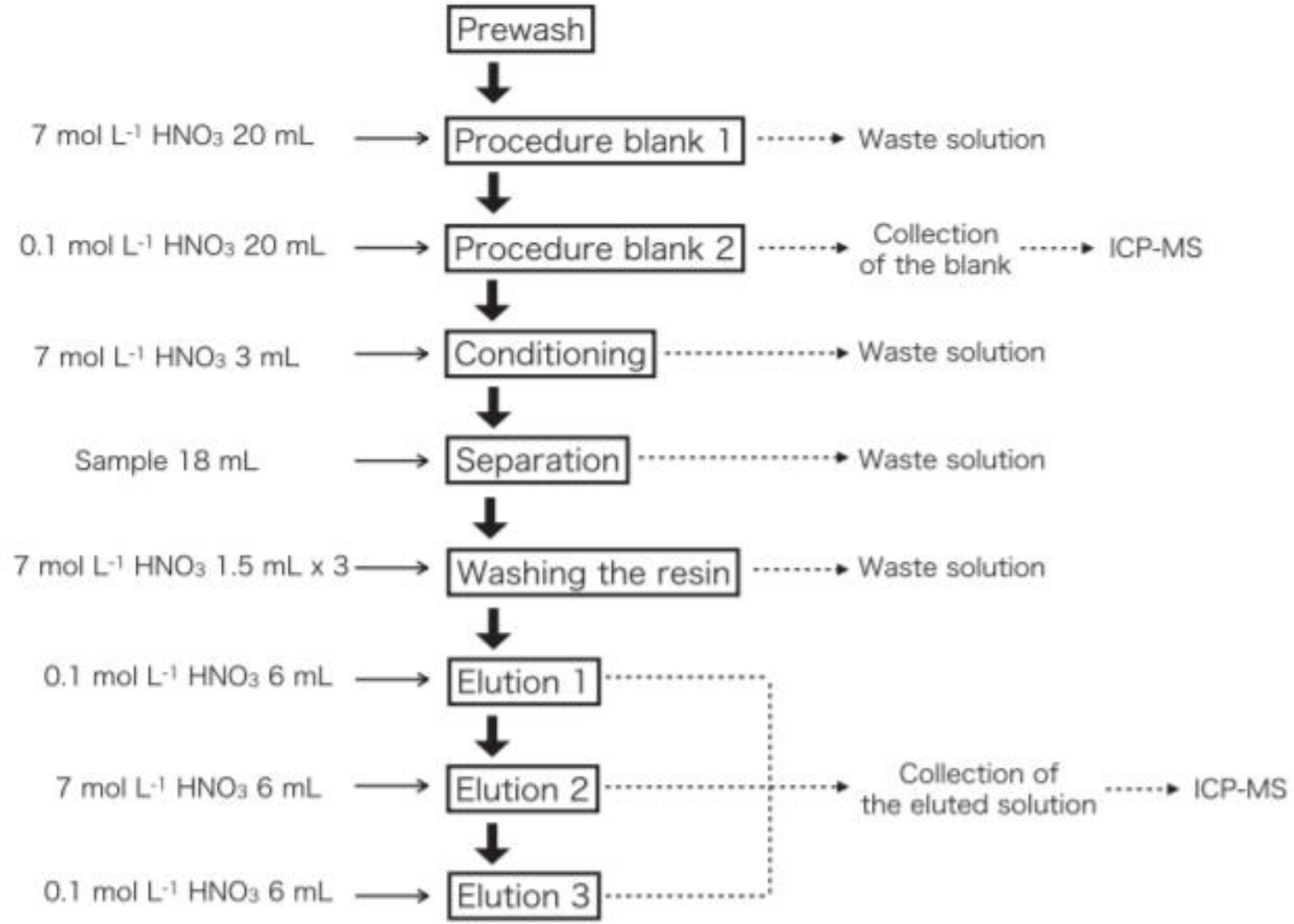
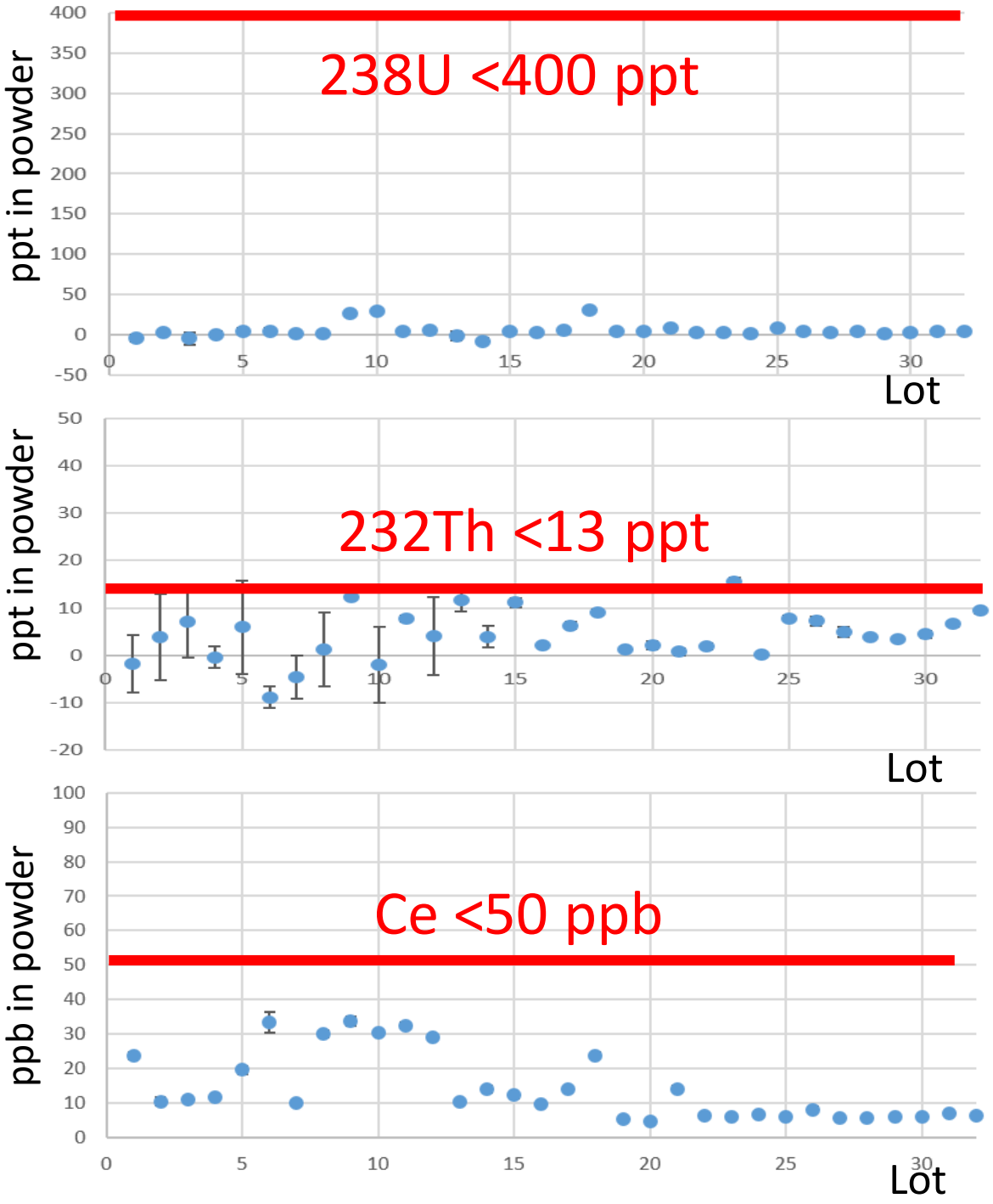


Fig. 6. Diagram of the whole procedure for the solid-phase extraction.


238U <400 ppt
ppt in powder
Lot
232Th <13 ppt
ppt in powder
Lot
Ce <50 ppb
ppb in powder
Lot