Artificial formation of alpha recoil tracks using an americium source

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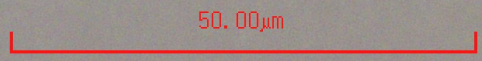
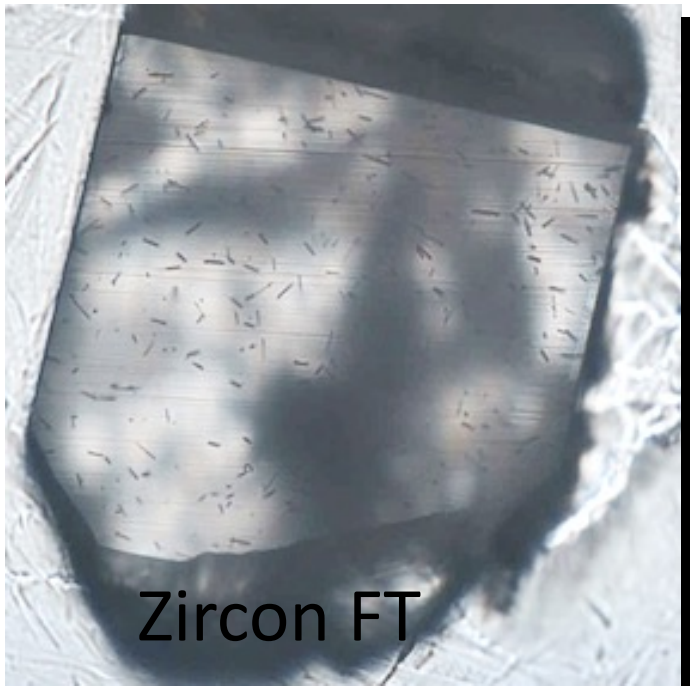


Research experience

- Geochronologist
  - Fission track dating
  - Luminescence dating
  - U-Pb Dating
  - K-Ar Dating
  - Quaternary environment (^{210}Pb, ^{137}Cs, ^{14}C)
- Re
- Active faults,
- Environmental fluctuations

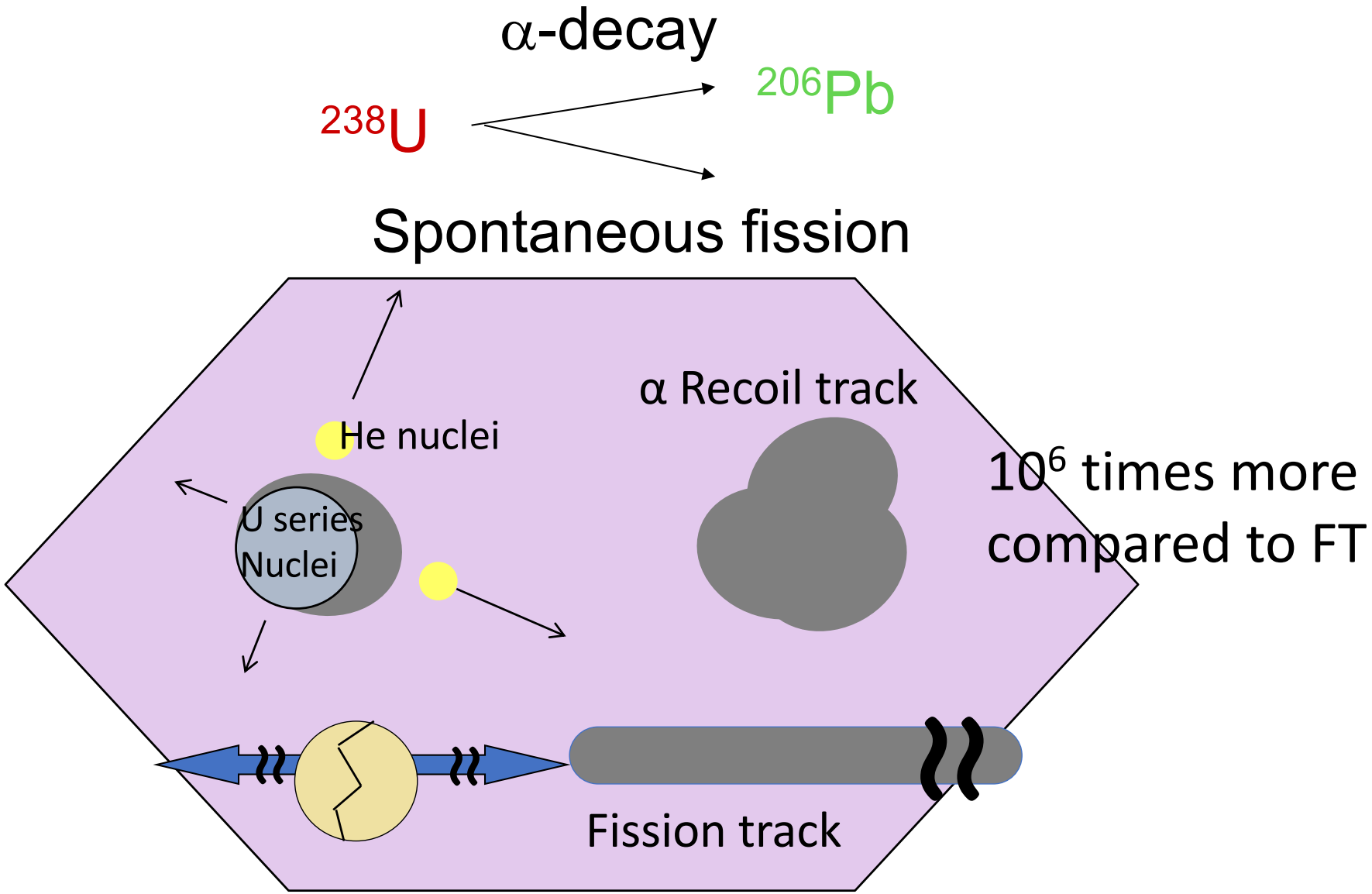
Dr. Hirose kindly invited me to this research community to understand space evolution

- Radiation from the Earth Surface Environment


Zircon FT


Feldspar Luminescence

Dating method by the observation of radiation tracks



Order of 100 yrs, 1000 yrs dating possible (zircon: 100ppmU)

Dating by radiation induced track counting

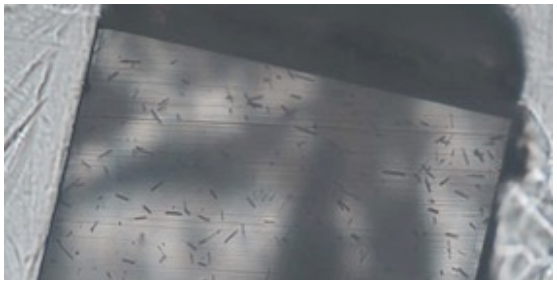
FT method is popular

	FT	ART
Product	Spontaneous fission of ^{238}U	Alpha dcaey of U, Th, etc.
Energy	~200 MeV	Several MeV
Number of tracks	few	10^6 times more compared to FT
Shape	width: 6~10nm length:15000nm (Apatite)	30~40nm (Biotite)
Annealing	Well studied (Ap: ~100° C, Zr: ~300° C)	???
Dating	FT method (matured) zircon, apatite, sphene, volcanic glass	ART method (On going) layerd silicate
Ref	e.g., Fleischer et al., 1975	e.g., Hashemi-Nezhad and Durrani, 1981

Order of 100 yrs, 1000 yrs dating possible (zircon: 100ppm U)

ART method?

Limit in applying FT dating

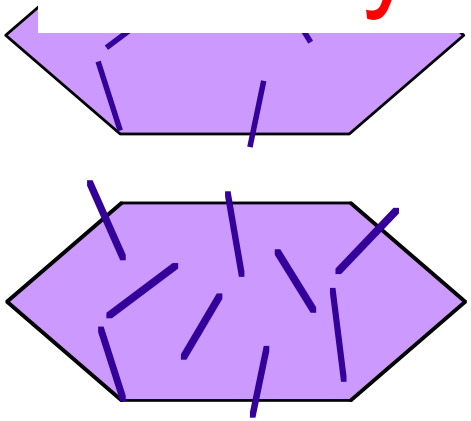


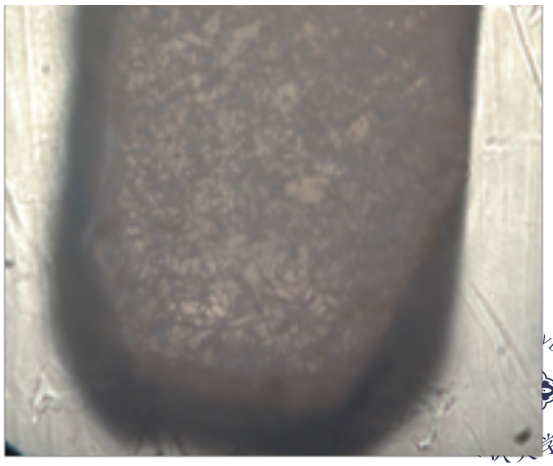
Countable zircon

Short etching time

High resolution microscope

Trial by Atomic Force Microscope





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Uncountable

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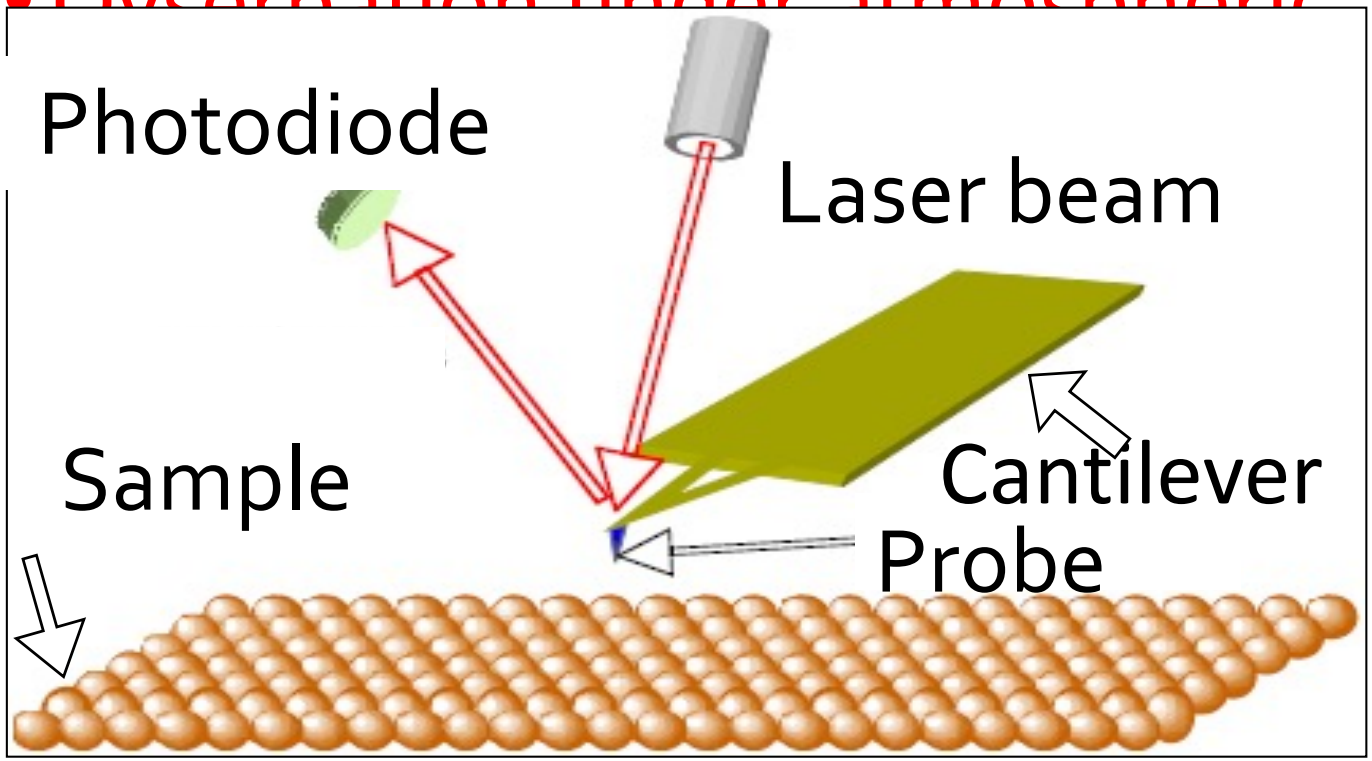
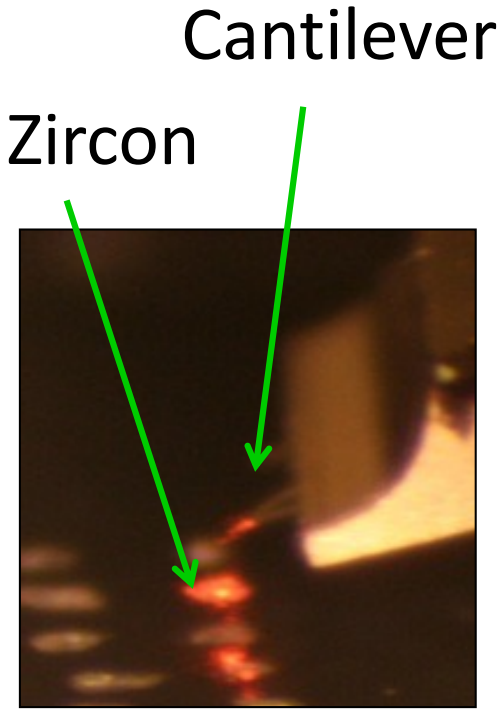
AFM (Atomic Force Microscope) Outline

- Scanning by a small needle
- No energy given ＝No FT annealing
- No carbon coating
- Oxserbation under atmospheric



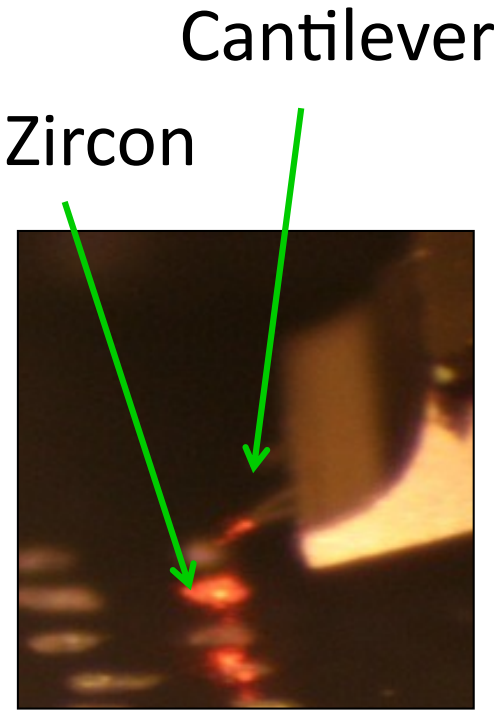


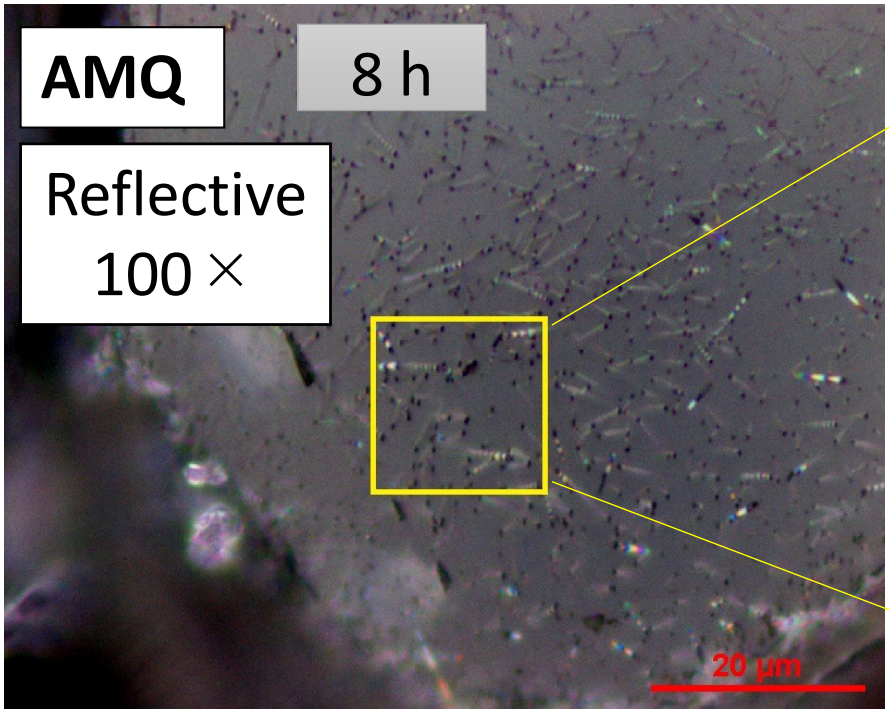
AFM (Atomic Force Microscope) Outline

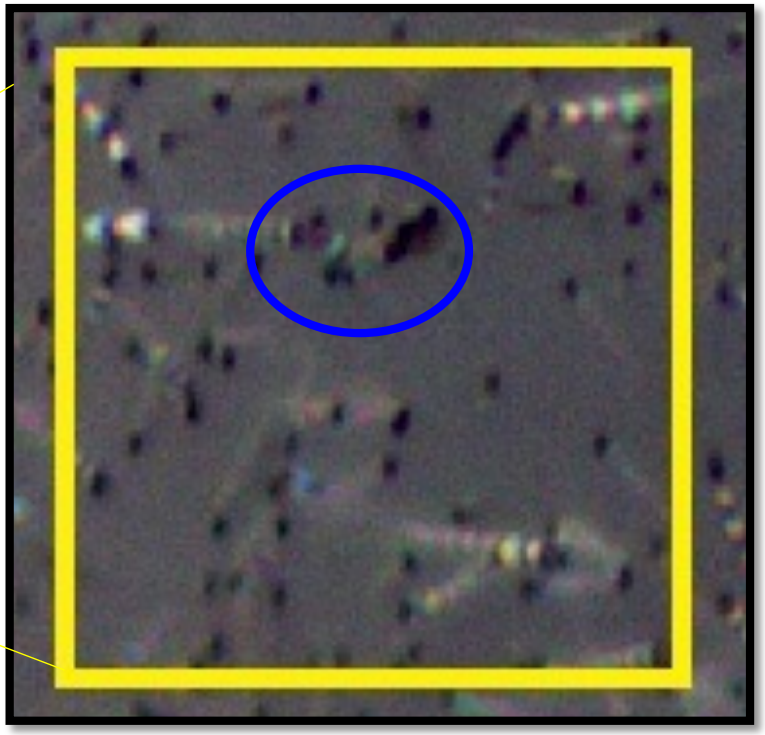
- Scanning by the small needle
- No energy given ＝No track annealing
- No carbon coating
- Observation under atmospheric pressure
- Depth profile of tracks (shallow)

By AFM

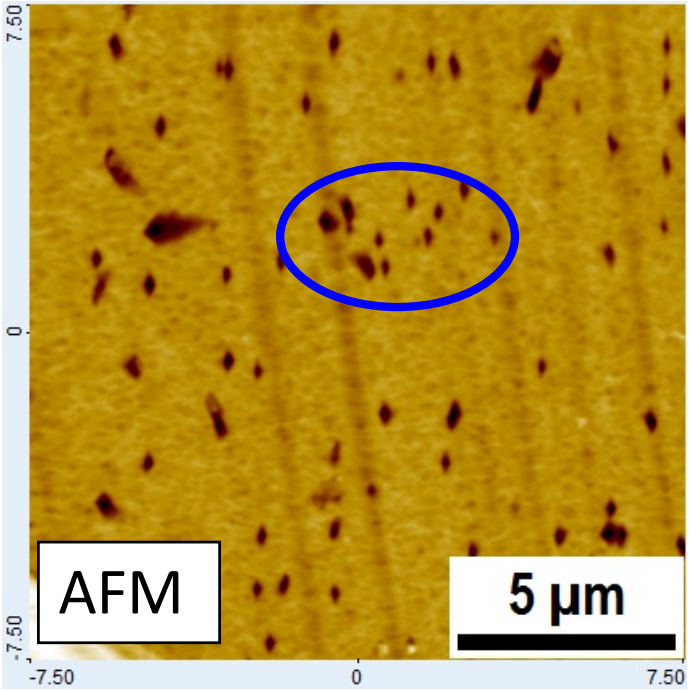
- nano-order observation
- short etching time
- no overlap of tracks



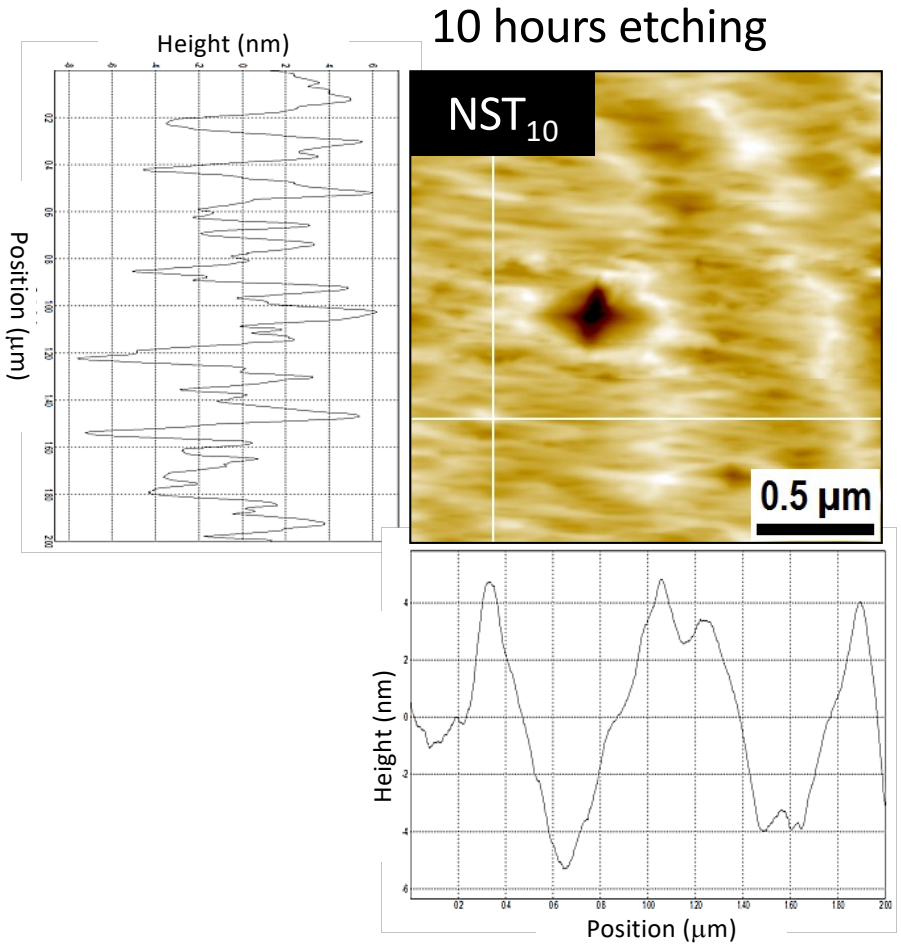


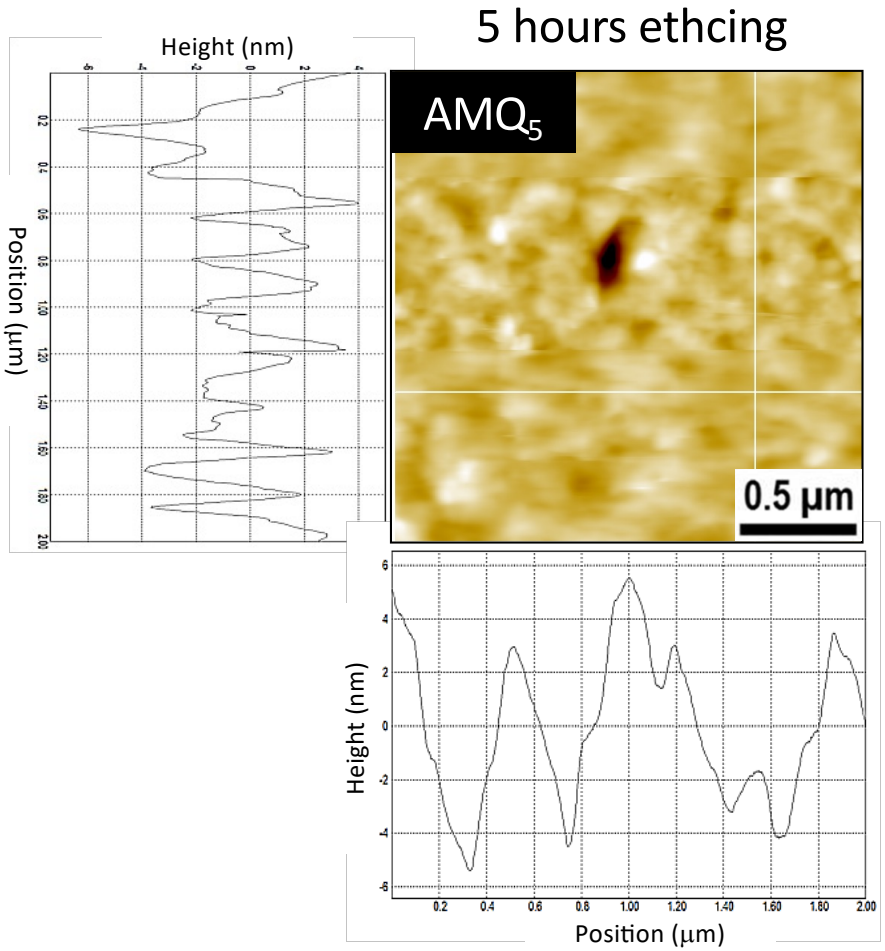


Tracks are connected under the optical microscope,
but distinguishable in AFM image.



Strange wavy structure



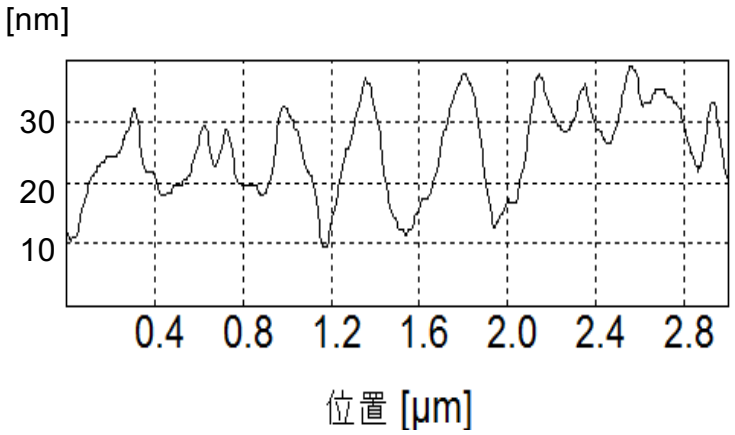


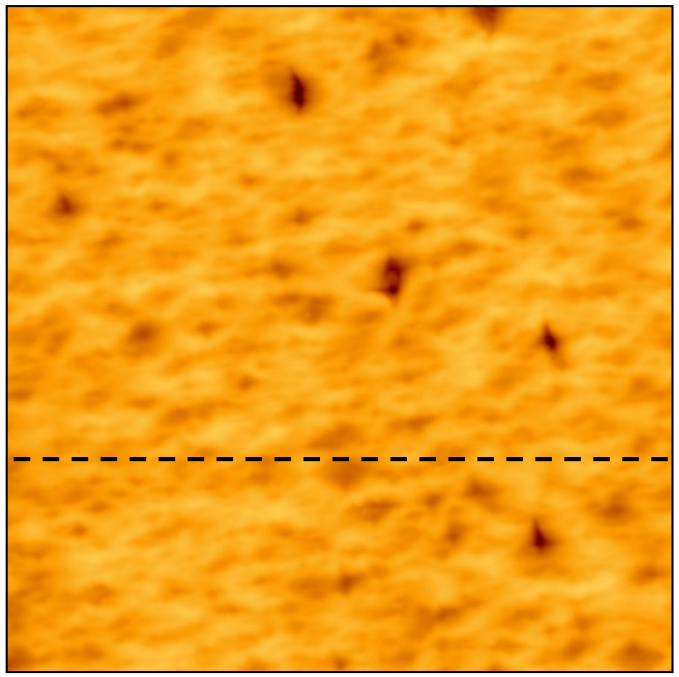
Amplitude: ~5nm (depth ~10nm)

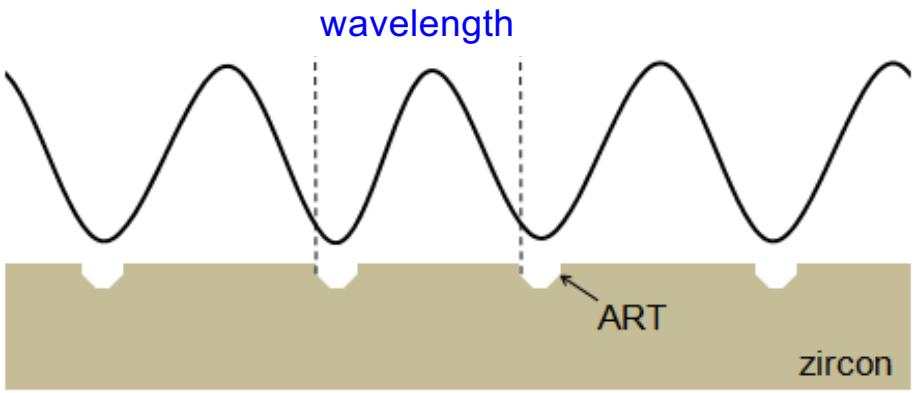
Ohishi and Hasebe (2011)

Surface structure: alpha recoil tracks?

☒ Rough estimate of number of ARTs from the wavelength — No match
ART estimate from FT age







Depth of pits:~25nm

- Topographic low at the ART
- Wavelength may reflect ART density

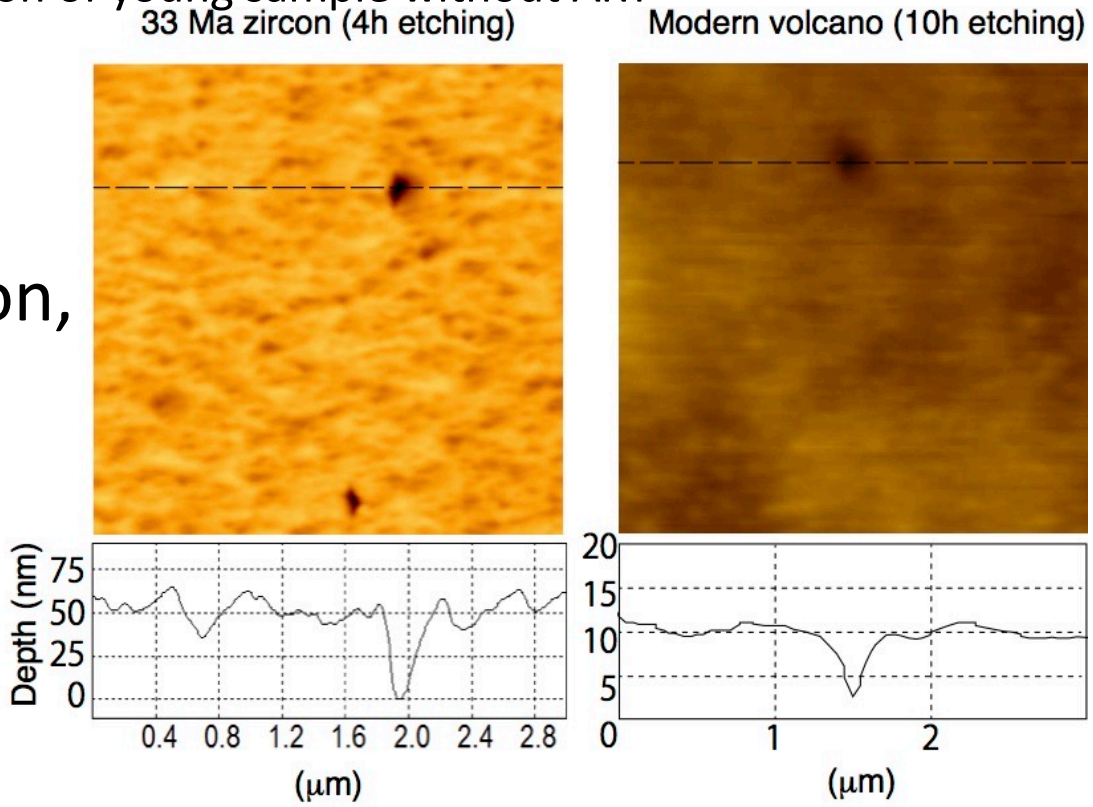
Rough estimate of number of ART

Surface structure: alpha recoil tracks?

☒ Rough estimate of number of ARTs from the wavelength
ART estimate from FT age
— No match

☑ Observation of young sample without ART

33 Ma zircon,
4h etcihing



Modern zircon,
10h etcihing

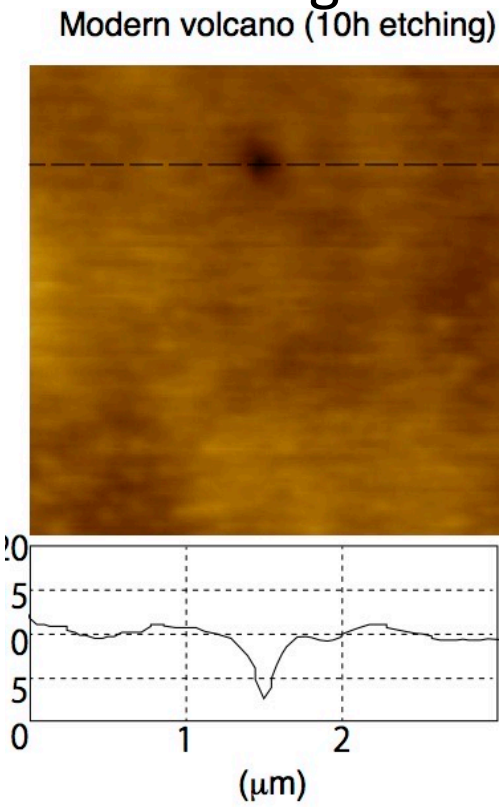
25nm oscillation
50nm depth FT

＜5nm oscillation
7 nm depth pit （ART?)

☑ Effect of annealing

Surface structure: alpha recoil tracks?

Modern zircon (No FT)
10h etching

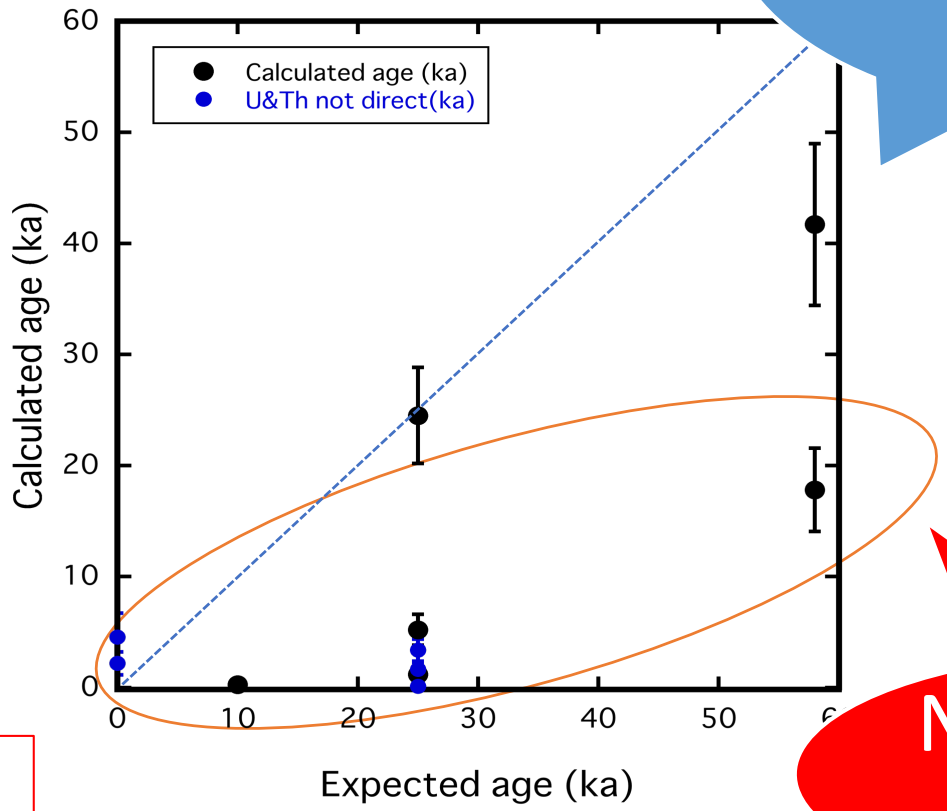


＜5nm oscillation
7 nm depth pit （ART?）

Unzen volcano

alpha recoil

Dating of age known Tephra zircon



(Hayasaka, 2018)

Not very successful

I need certificate they are ART

More haste, less speed

○**How we can produce artificial ART?**

✓ U size heavy ion irradiation necessary

RIKEN ($_{113}$Nh)
....hesitate to access...

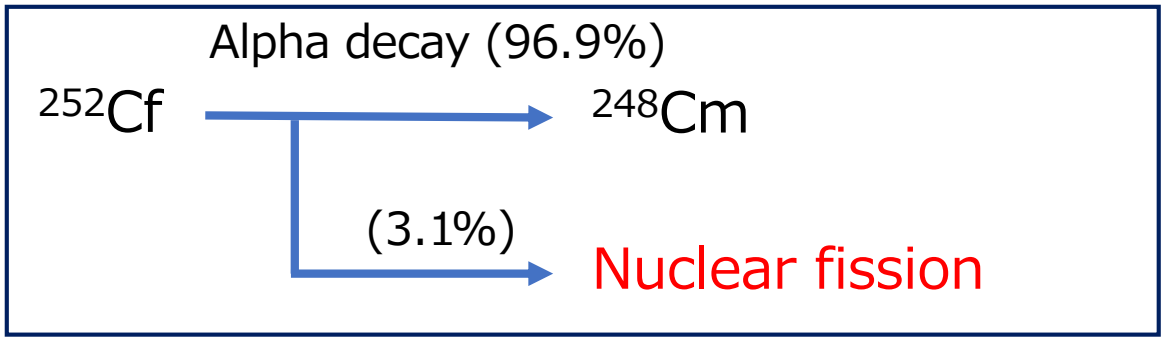
✓ Many irradiation facility: Positively charged ion

Alpha particle (He^{2}+), then remaining parent particle negatively charged

(Kobayashi and Oka, 2011)

Artificial ART: previous research

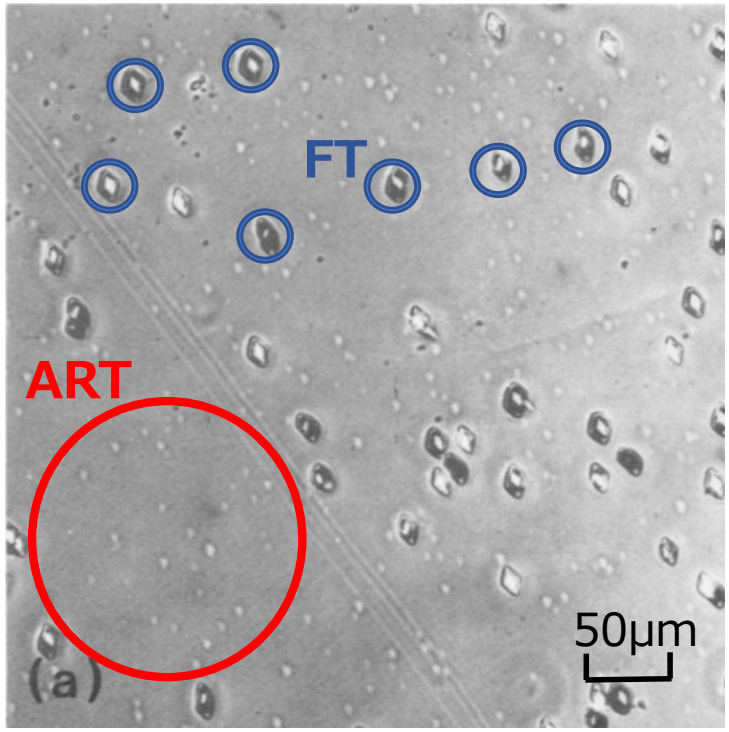
Hashimoto et al.(1980)
→ ^{252}Cf source (irradiation time 1~60min, under vacuum 0.01torr)



Disadvantage

- ART and FT formation
- FT noise and sputtering (pollution and uneven track distribution)

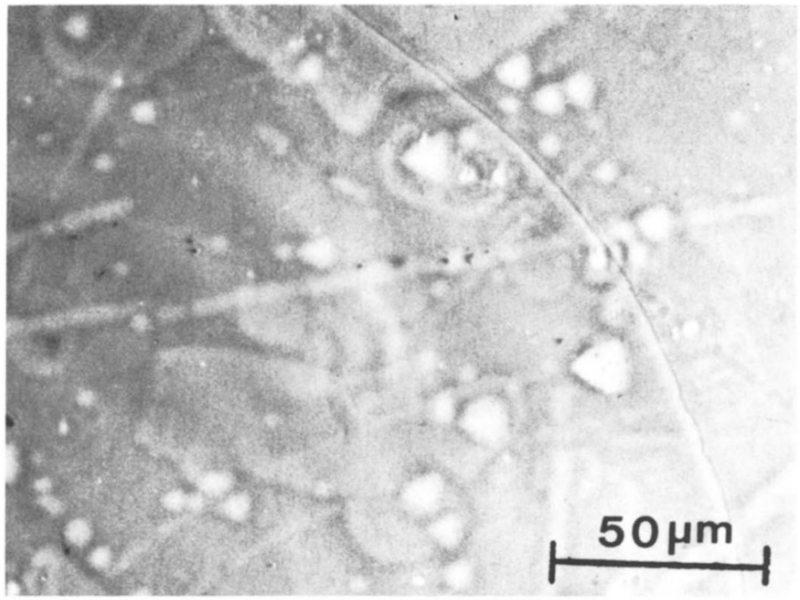
Source without fission decay



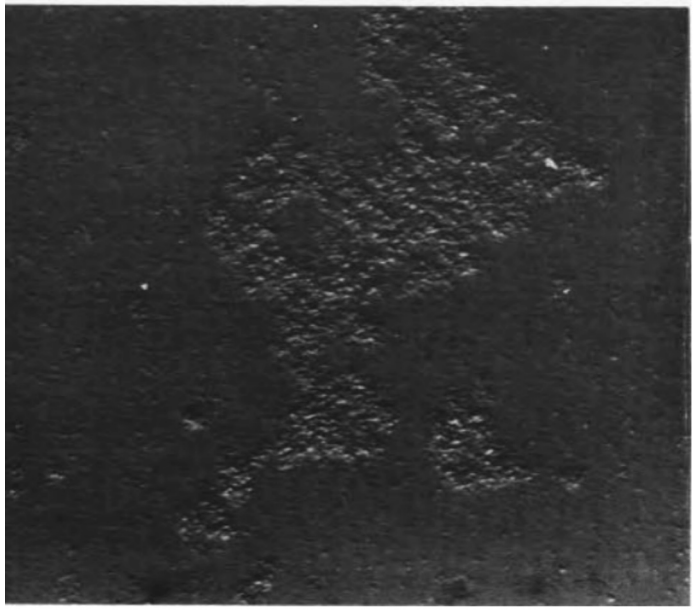
(Hashimoto et al., 1980)

Artificial ART : previous research

Turkowsky(1969), Hashemi and Durrani(1981)
→Drop of ^{241}Am or ^{228}Th solution on the sample surface



(Hashemi and Durrani,1981)



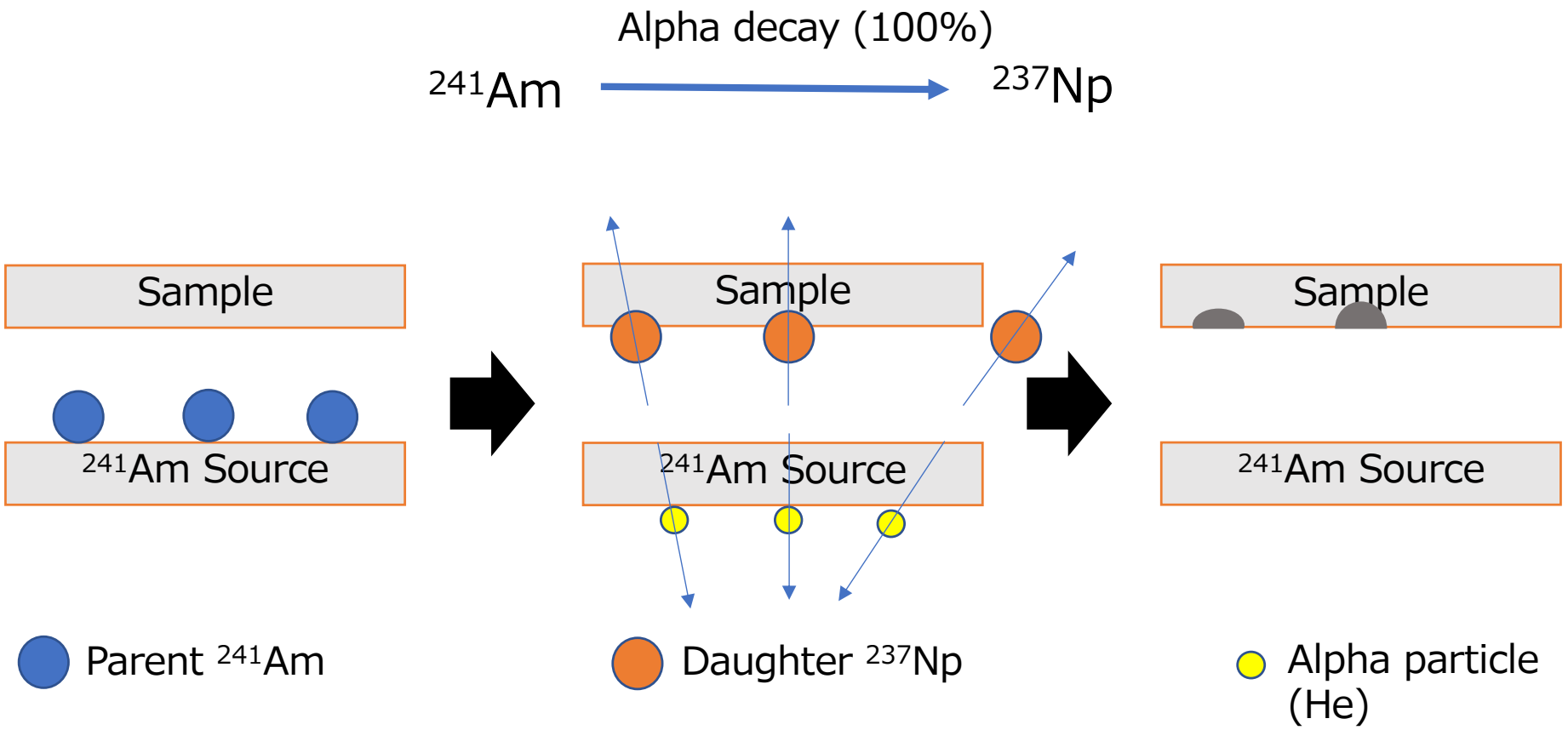
(Turkowsky, 1969)

Disadvantage

- pollution
- uneven ART distribution

We tried metal deposited ^{241}Am source

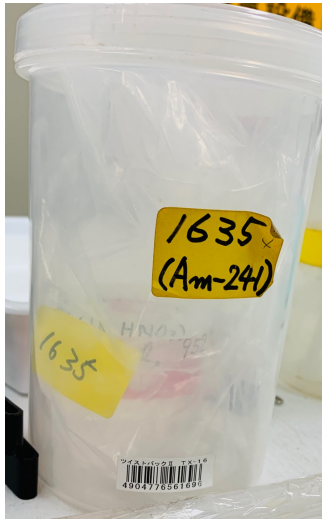
ART formation

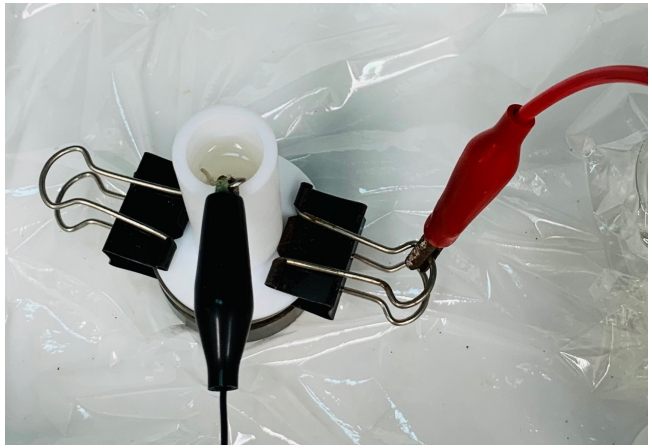


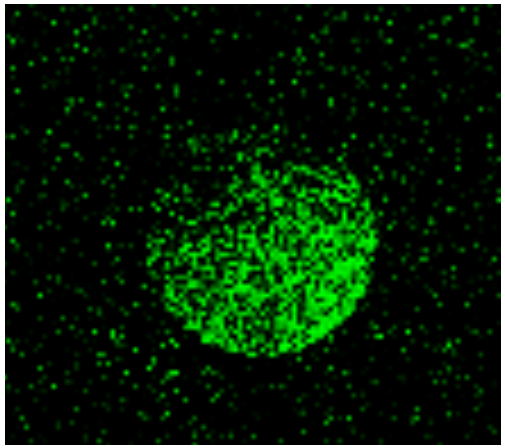
Sample: Muscovite
Am source: ??

^{241}Am Source with appropriate dose

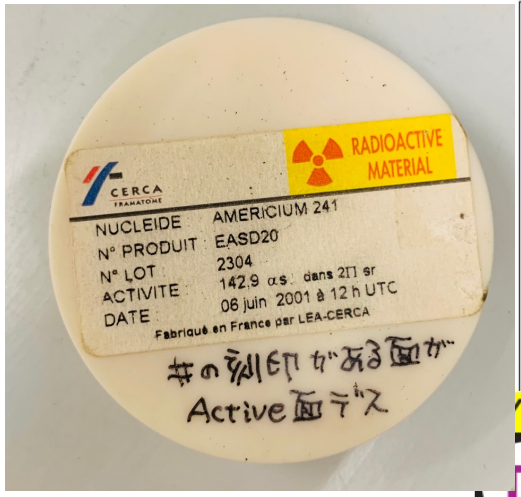
Electrodeposition: No success (uneven)

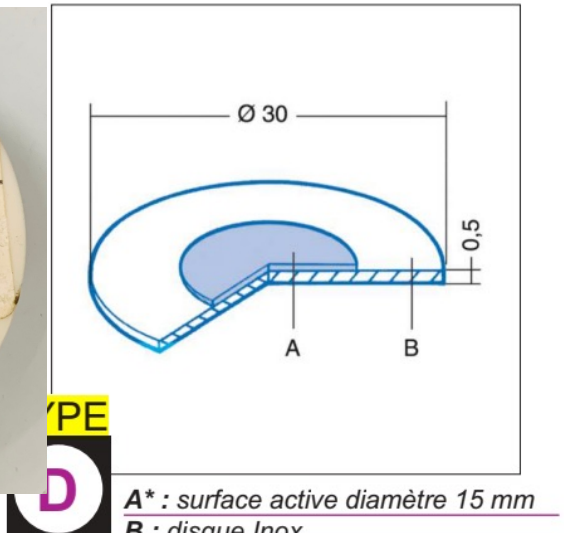






Commercial source

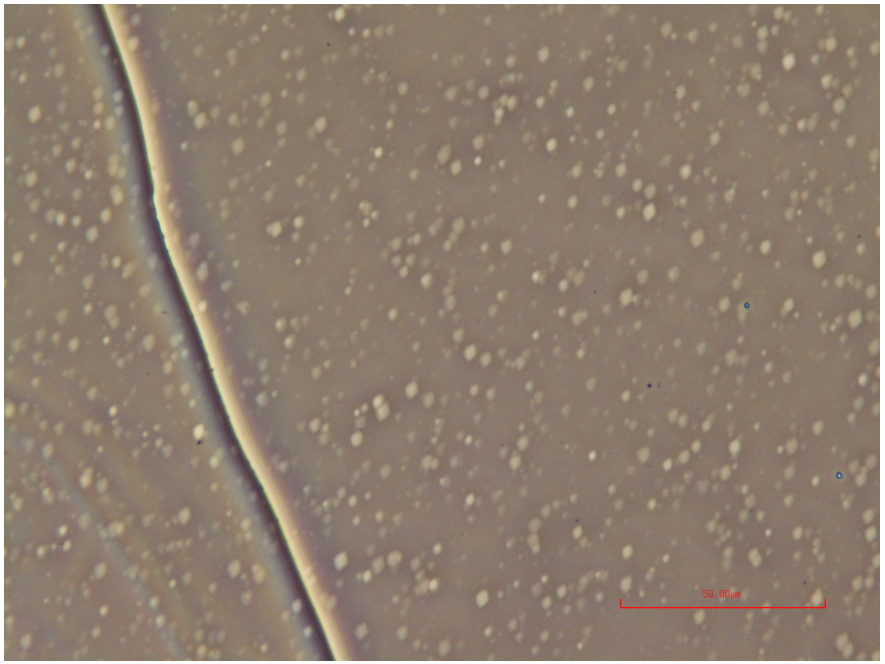




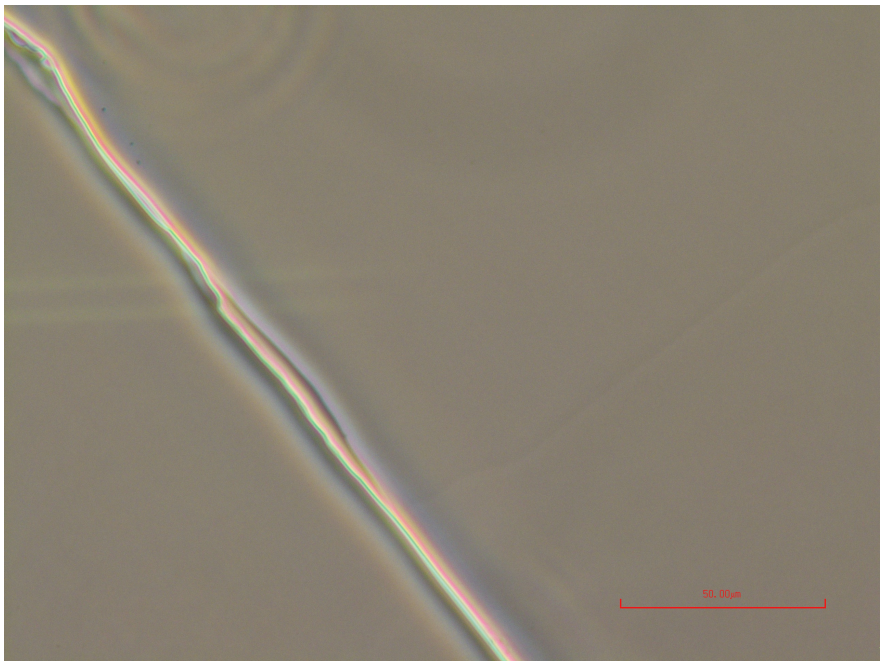
Base	Stainless
Radioactivity	300Bq
Method	Electrodeposition

Natural ART-like tracks in Muscovite

Before annealing



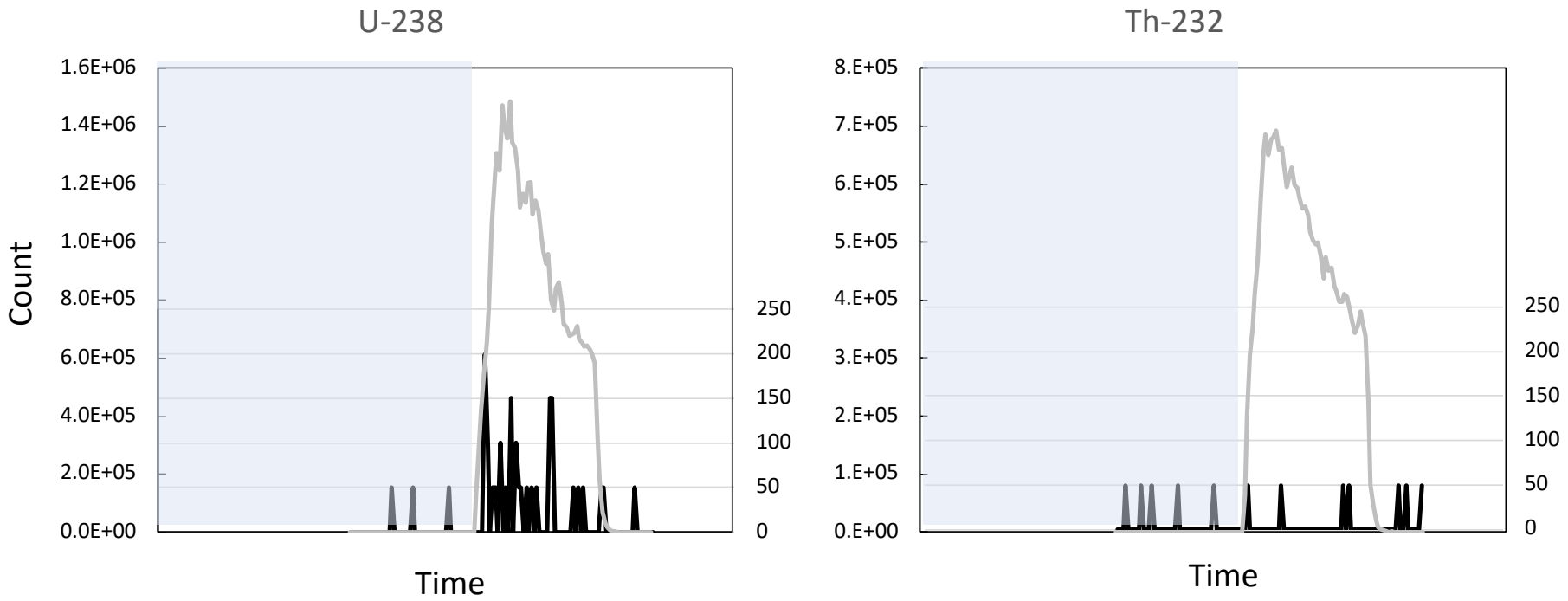
After annealing (600℃, 6h)



160 μm

Background tracks were all erased.

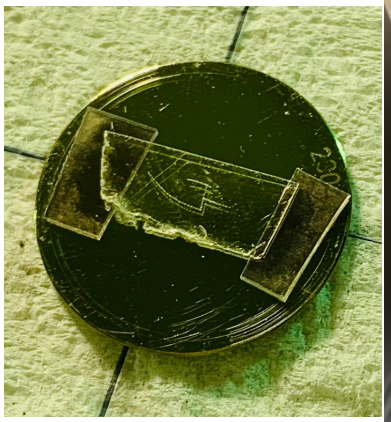
Uranium and Thorium concentration

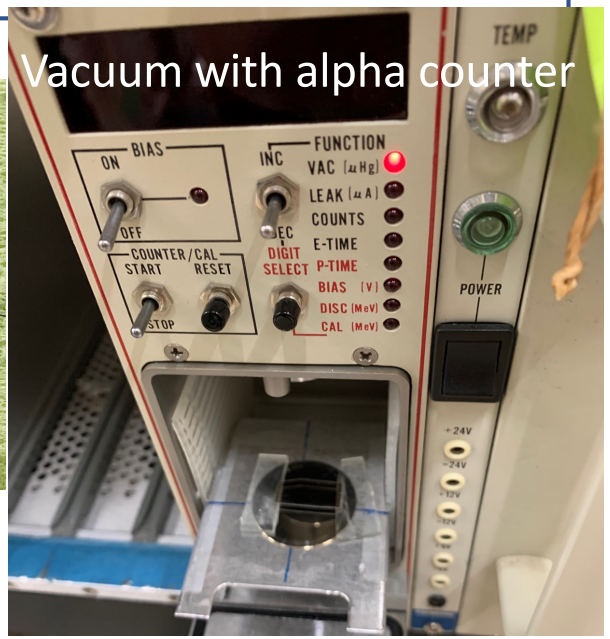


Irradiation and observation

①Pre annealing	600℃ 6 hrs (Hashimoto et al., 1980)
②Irradiation	^{241}Am (300Bq) 1hr 3hr 6hr 12hr 2 d 4 d 1 w
③etching	32℃ HF 2 hr (Gogen and Wagner, 2000)
④observation	Phase contrast microscope, ImageJ

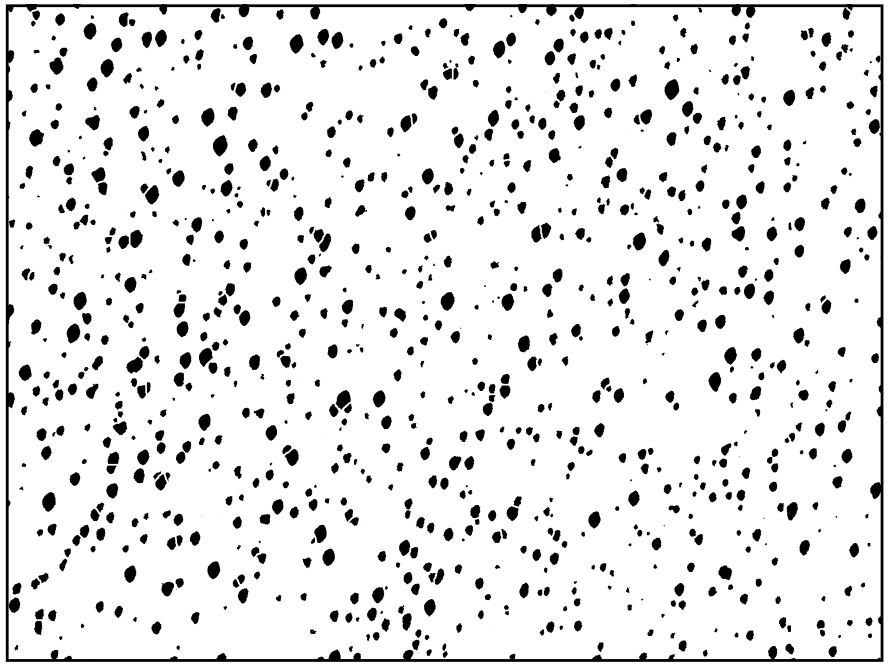
- Photo of 5 screens (34,100μm^2×5)
- Image binarization using ImageJ
- Number and size (area in μm^2) of ART





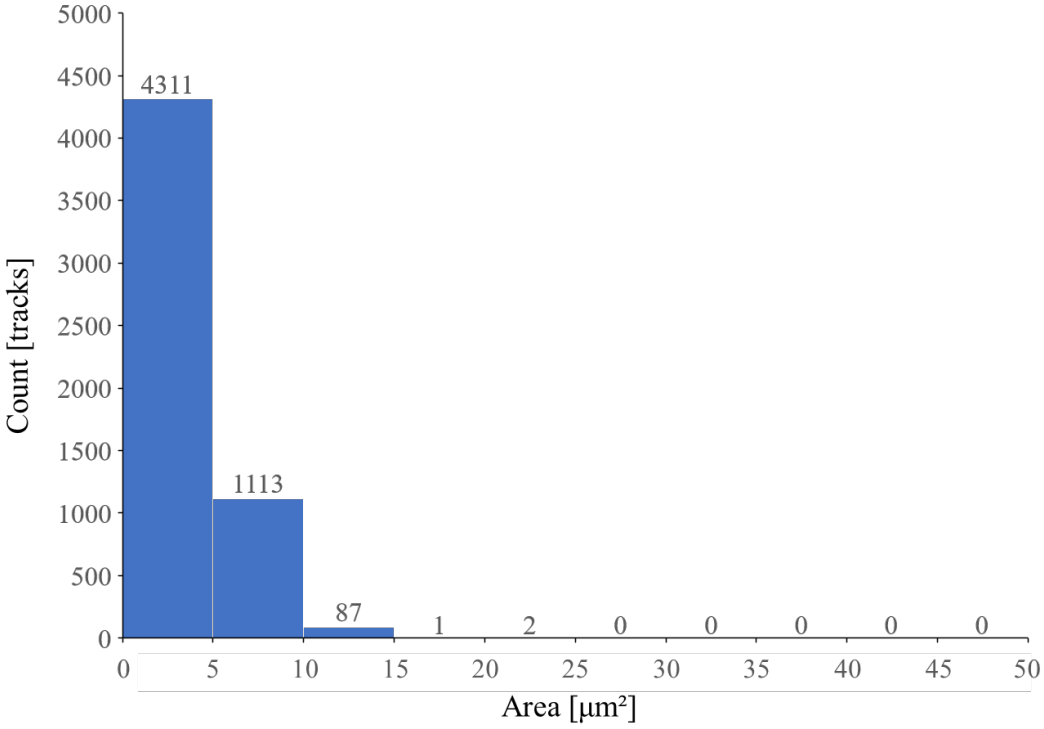
Background tracks

ImageJ



Number of tracks (5 screens) : 5516

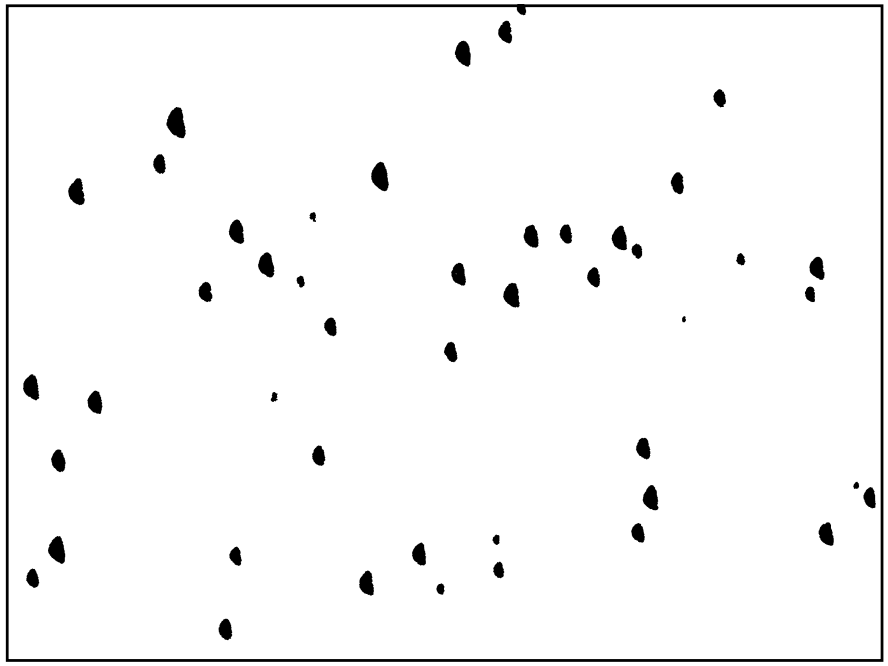
Size distribution



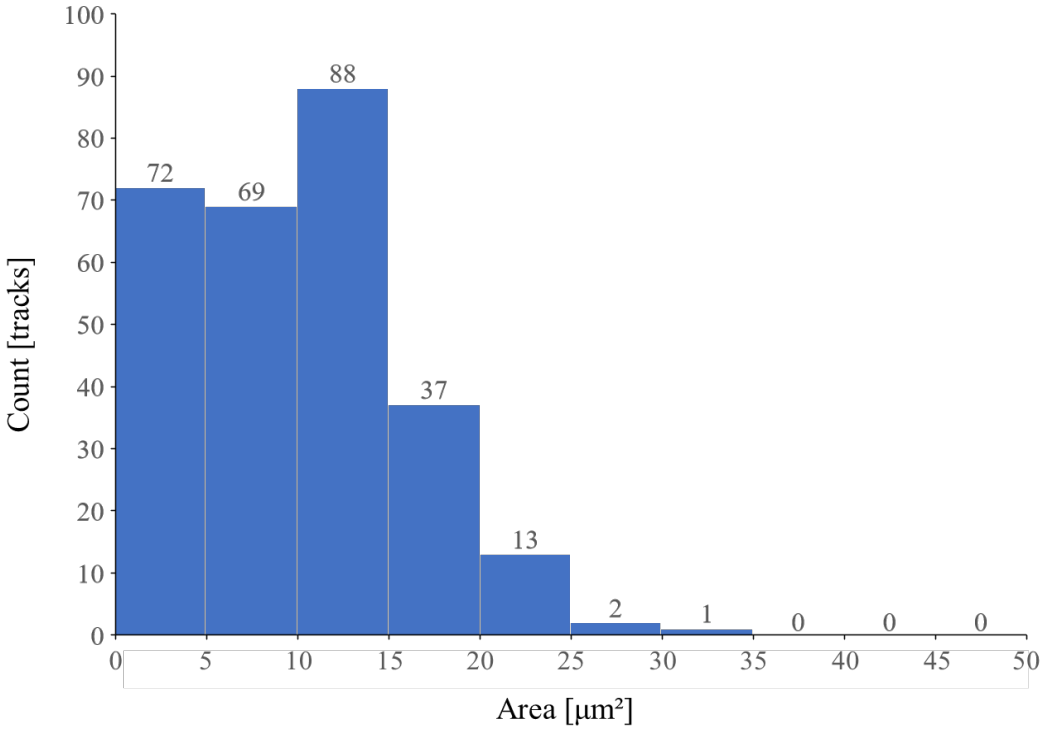
(0~5μm^2 tracks: around 78%)

After 1h irradiation

ImageJ



Size distribution



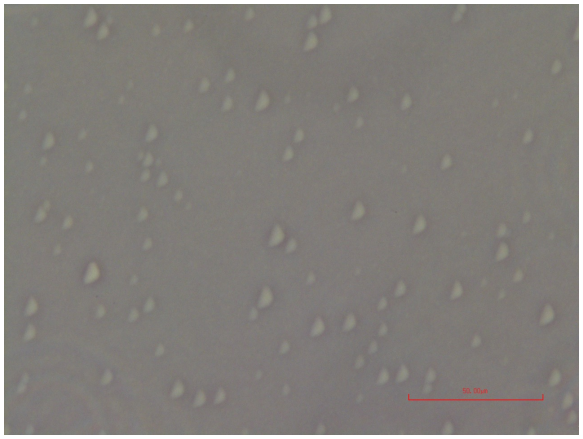
Number of tracks (5 screens) : 283

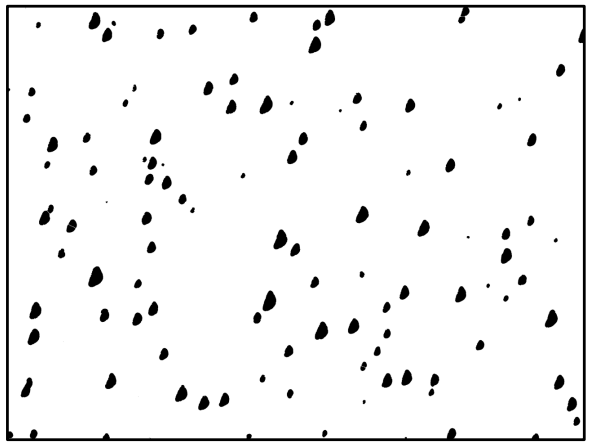
ART formed on the surface

→ARTs are bigger than natural background tracks

Results (3hr, 6hr, 12hr)

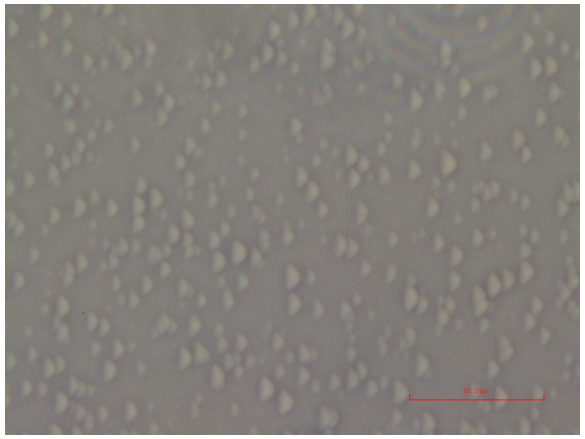
(3hr)

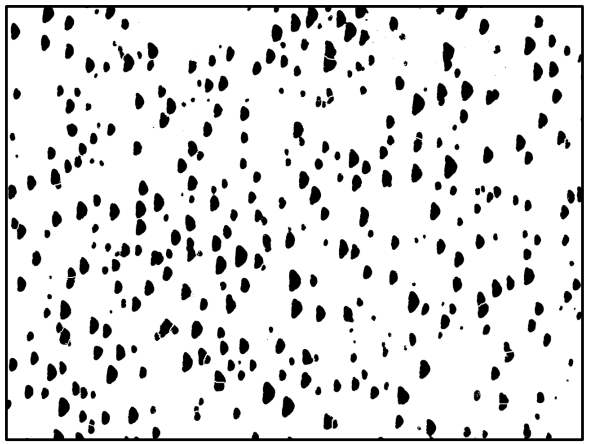




783 tracks

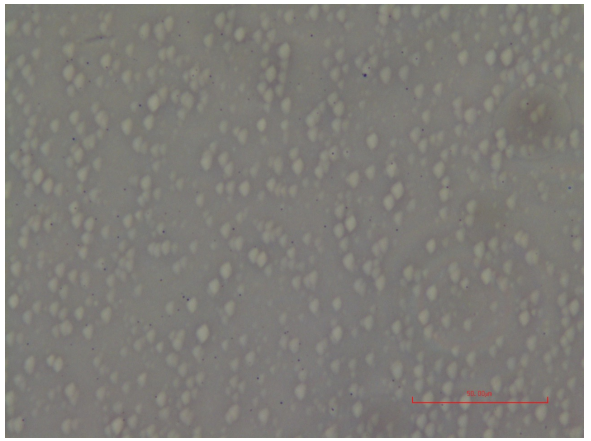
(6hr)

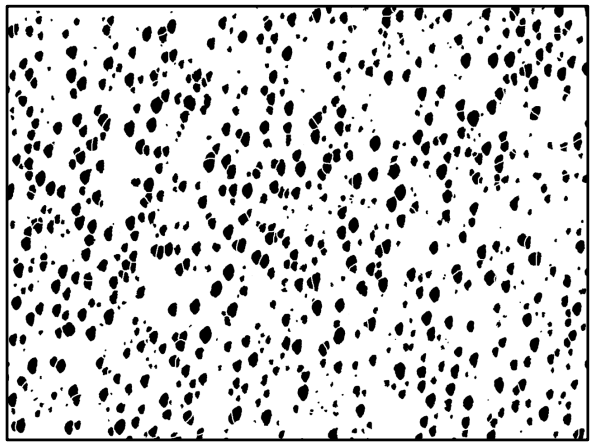




1959 tracks

(12hr)



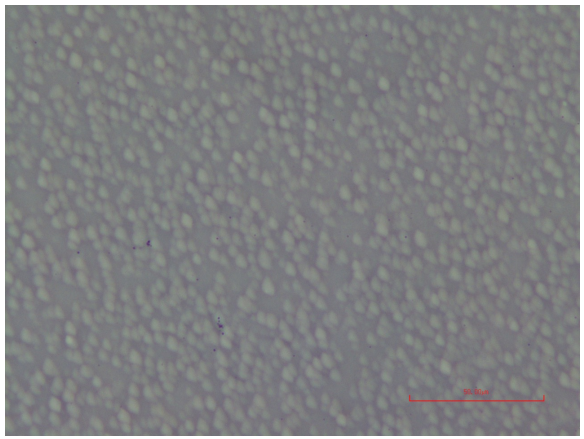


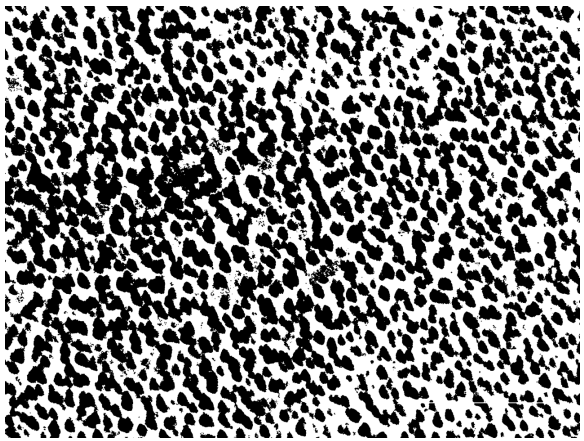
3666 tracks

Irradiation time increase→ Number of ART increase

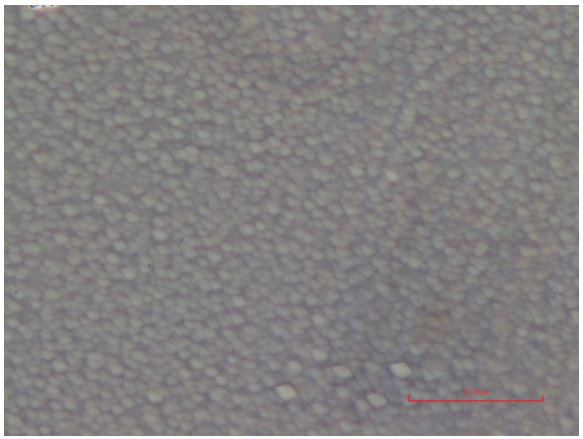
Results (2d, 4d, 1w)

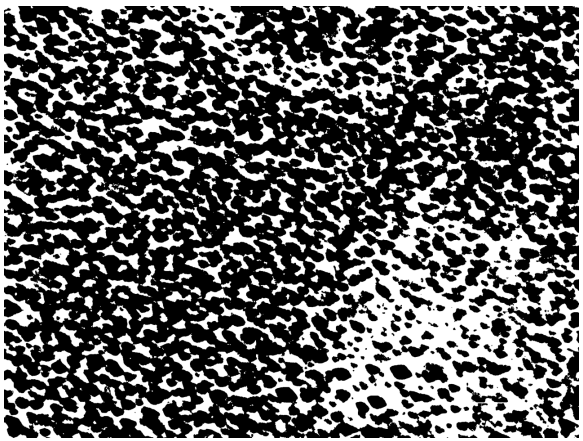
(2 days)



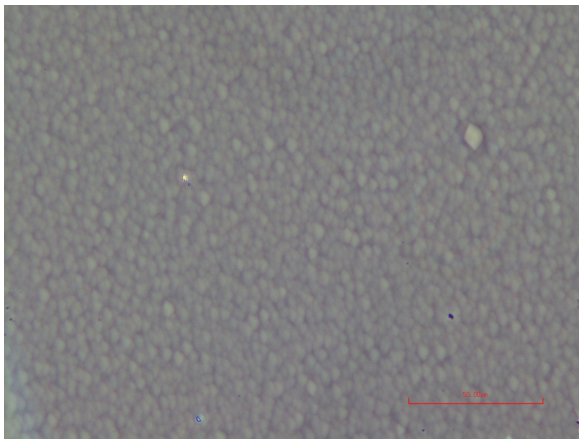


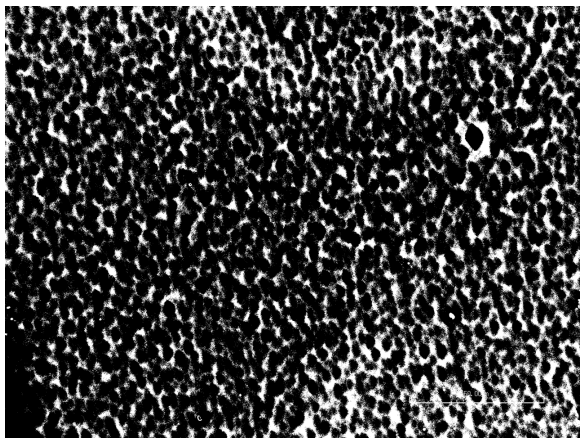
(4 days)





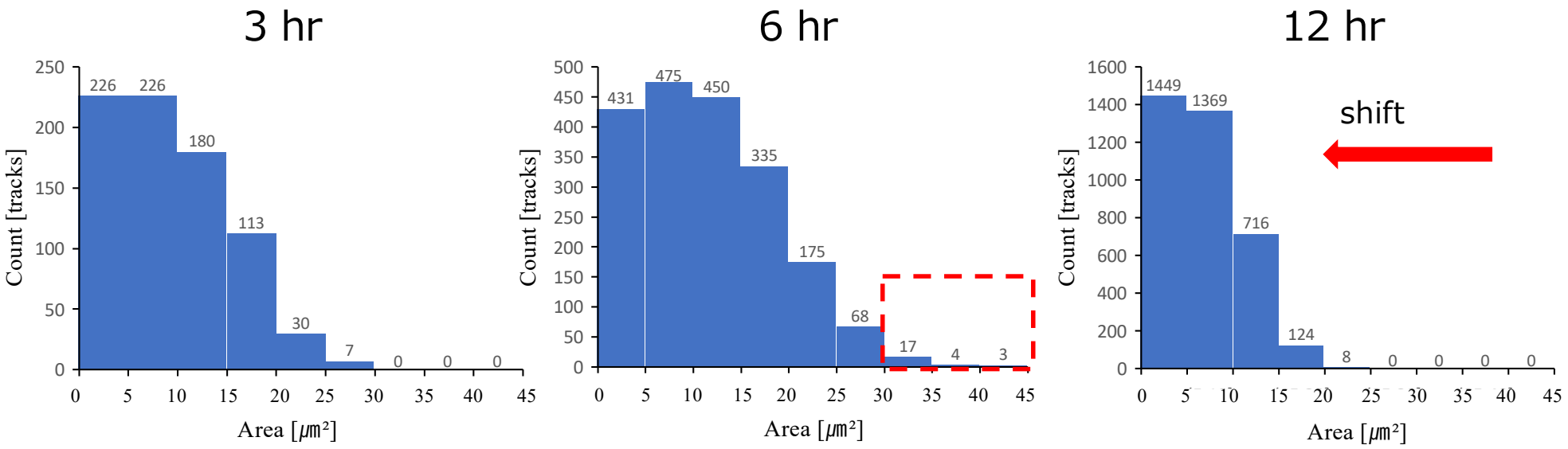
(1 week)





ART overlapping, unable to distinguish

Size distribution (3hr, 6hr, 12hr)



3 hr → Similar distribution to 1hr irradiated sample

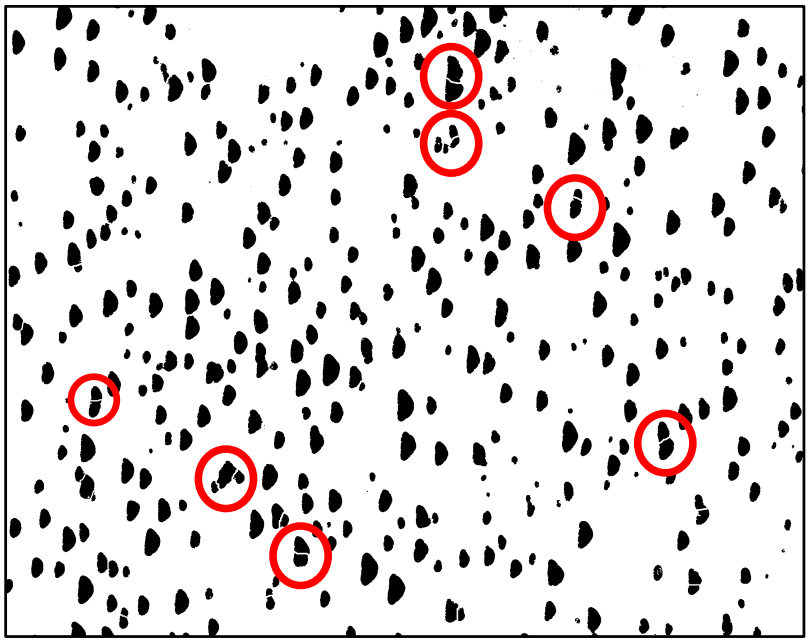
6 hr → larger tracks (30~45μm^2) are observed

12hr → distribution sift to left (in 0~20μm^2)

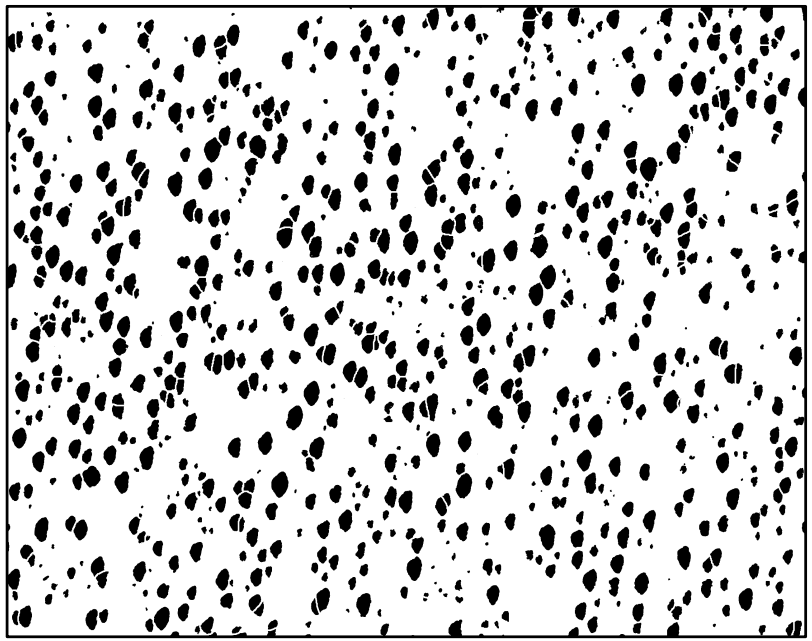
Overlapping of ART affects the apparent size distribution

ART overlapping

6時間



12時間



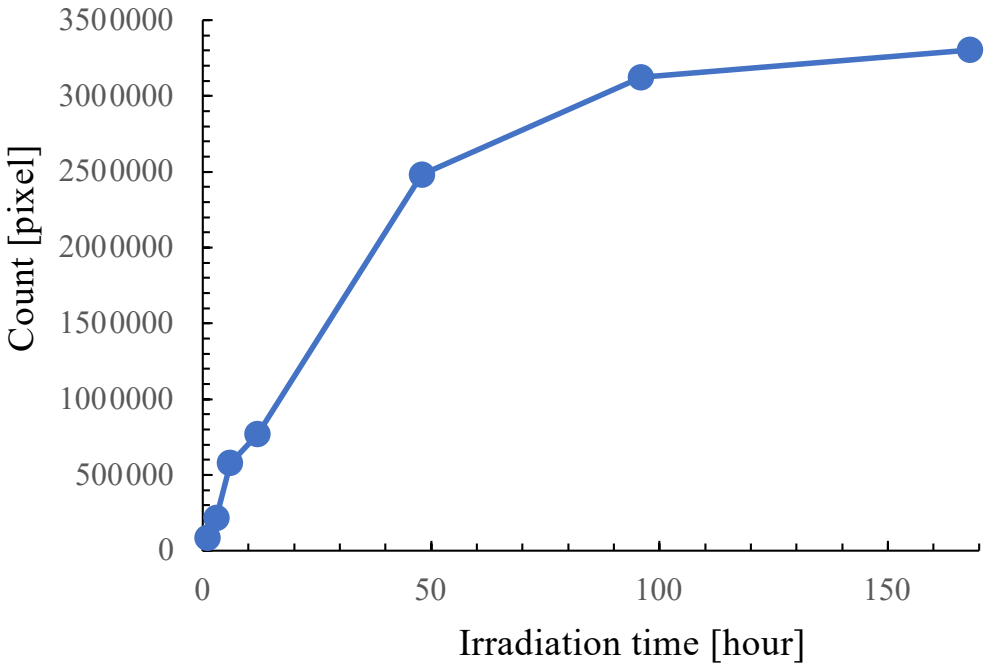
→ Manually separated based on original microscope image

→ Then they are smaller than original size

→ When overlooked, two ART combined and they are larger.

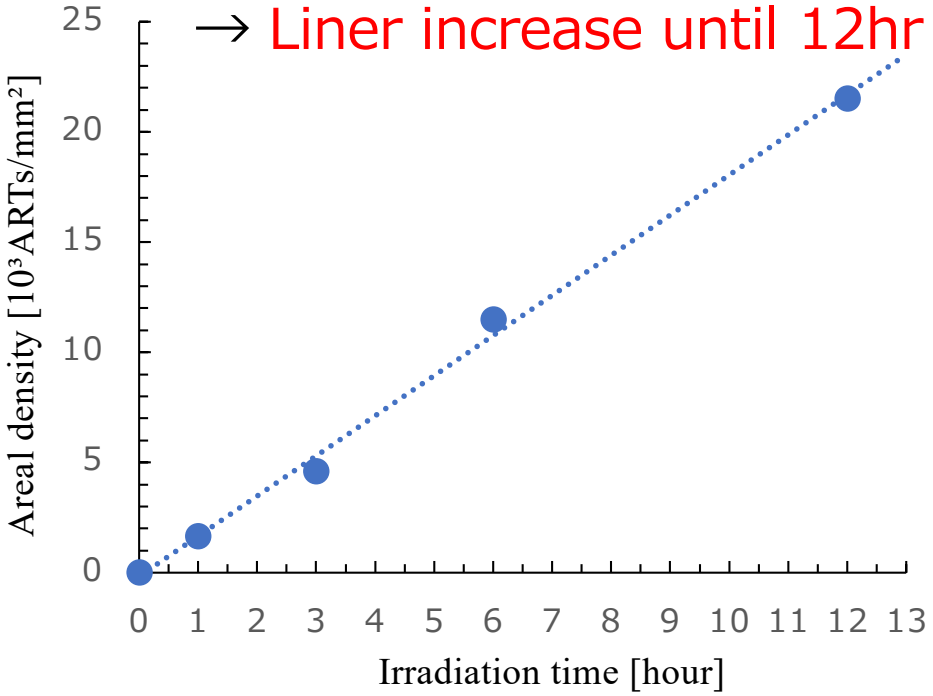
Number of ART vs irradiation time

Time vs pixel number



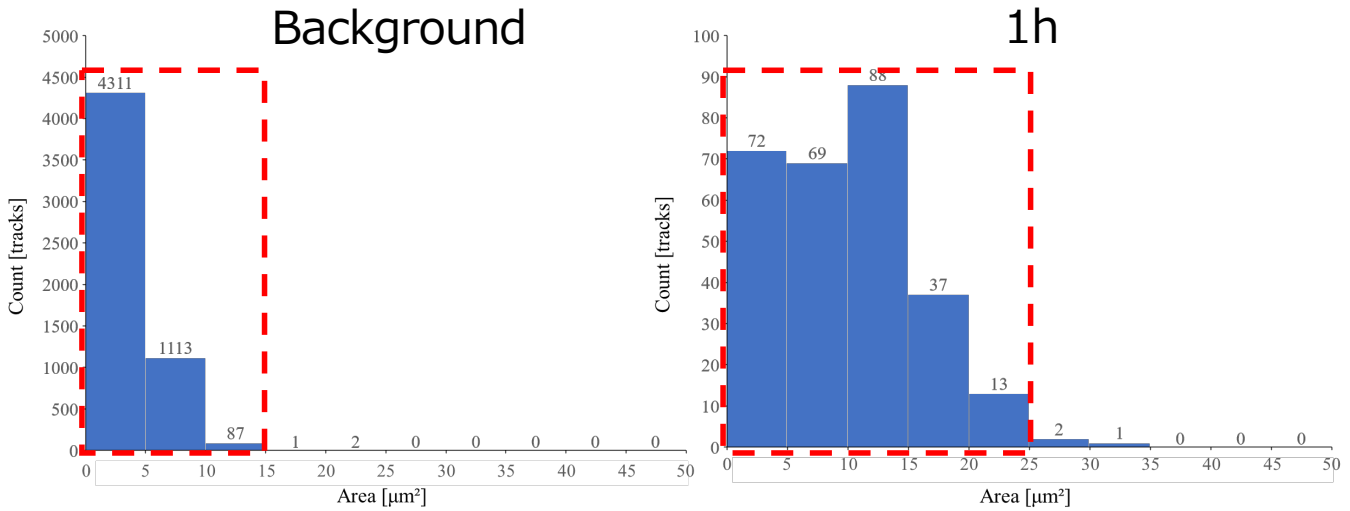
Time vs ART areal density

→ Liner increase until 12hr



Giving a limit in dating under this etching and observation condition

Comparison between Artificial and Natural



Number of alpha decay

Natural (1~8) > Artificial (1)

Average ART size

Natural (0~15μm^2) < Artificial (0~35μm^2)

Long-term annealing at ambient temperature?

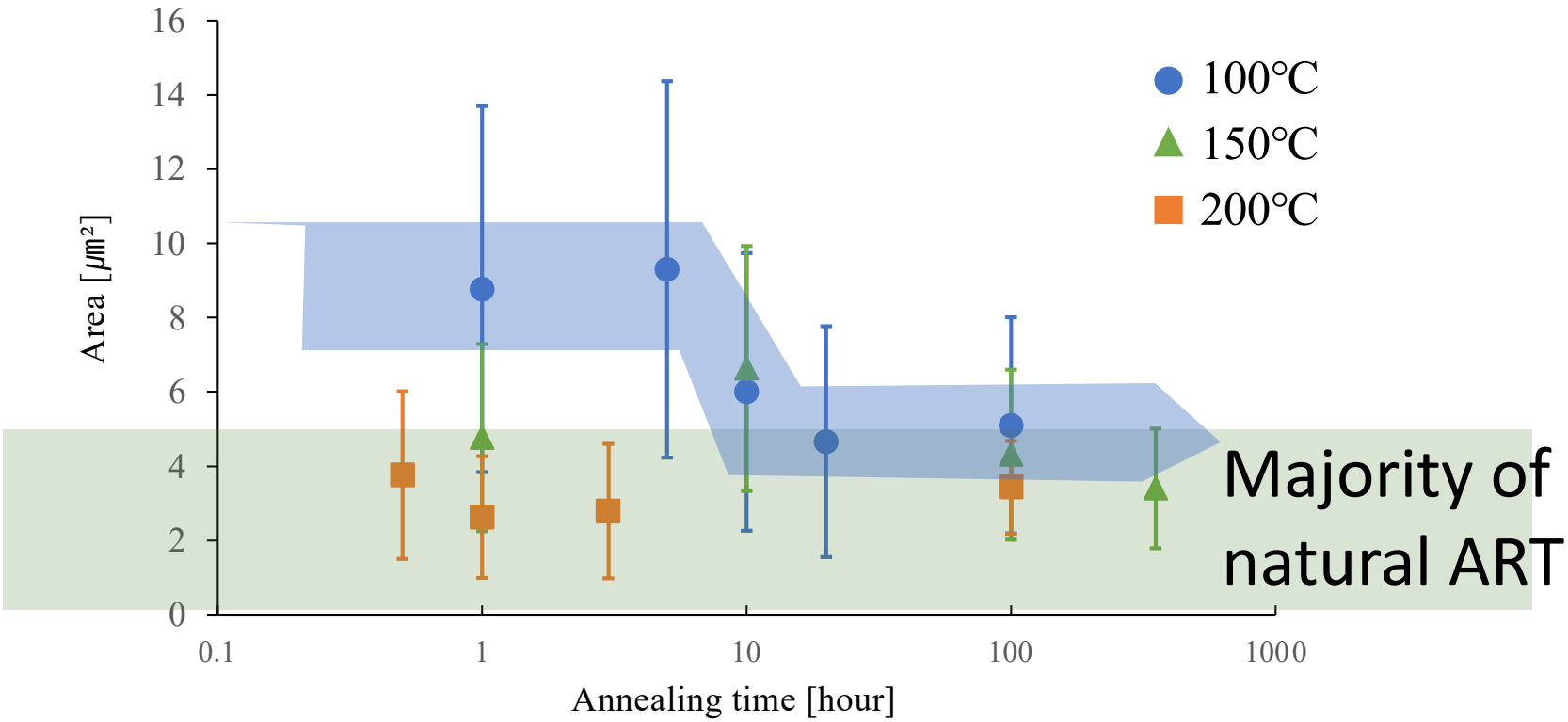
⇒ annealing experiment

Annealing behaviour

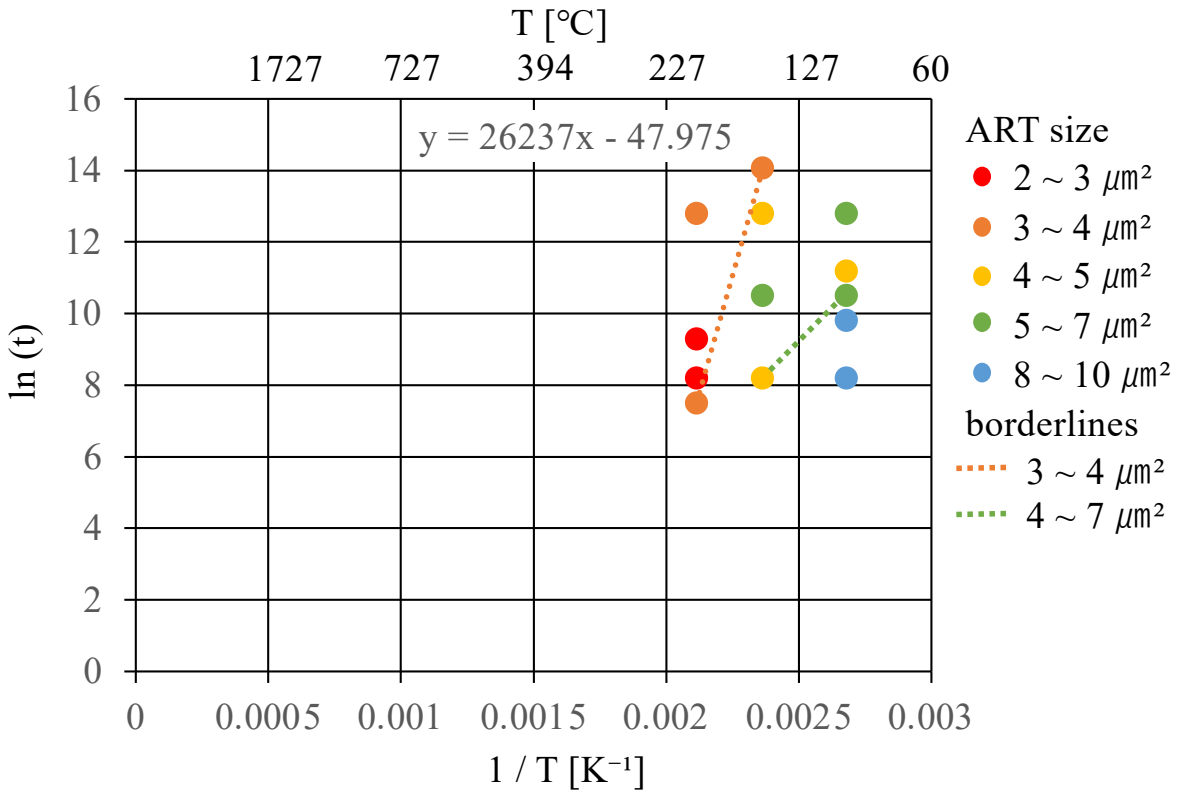
・Preannealing →3hr ^{241}Am irradiation →Annealing

Temp : 100℃,150℃,200℃

Time : 30m,1hr,3hr,5hr,10hr,20hr,100hr,352hr



Geological annealing duration



Annealing duration

Temperature (℃)	Estimated period (Gyrs)
25.0	8.0
26.0	6.0
27.0	4.4
28.0	3.3
29.0	2.5
30.0	1.9
31.0	1.4
32.0	1.1
33.0	0.8
34.0	0.6
35.0	0.5
36.0	0.4
37.0	0.3

→Long tern annealing at ambient T (27℃~34℃) can shrink the track size

Little size variation → recent heating at the time of sampling? They may not be ART?
Internal ART vs surface ART

Conclusion

Positive

Artificial ART formation successful.

ART can be a tool for thermochronology.

Negative

Not truly the same situation with natural ART

(only from crystal outer surface, only one decay)

Question arises

What we find in natural mica?

Result and discussion

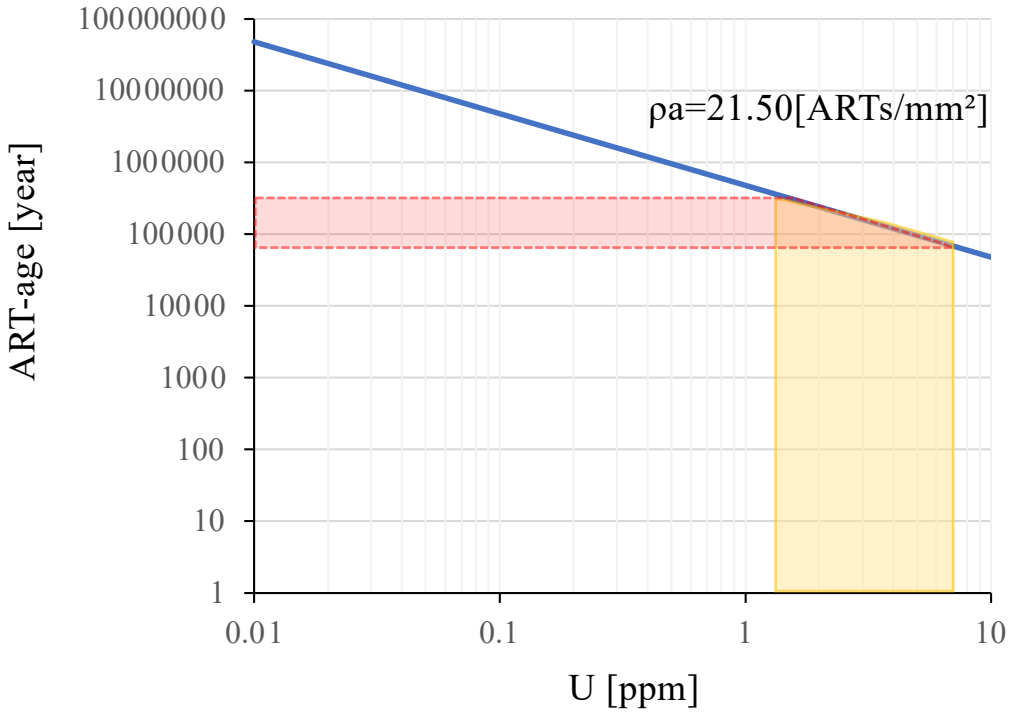
○ART dating applicability to muscovite (2h etching)

ART age equation (Gogen and Wagner, 2000)

$$t = \frac{\rho_a}{T \cdot \left\{ {}^{238}U_a \cdot \left({}^{238}\lambda + {}^{235}\lambda \cdot I + {}^{232}\lambda \cdot \left[\frac{Th}{U}\right]_s \right) \right\}}$$

Age: t — ART areal density: ρ_a — Diameter of ART: T — U concentration: ${}^{238}U_a$ — Decay constant: ${}^{238}\lambda$, ${}^{235}\lambda$, ${}^{232}\lambda$, I — Th/U: $\left[\frac{Th}{U}\right]_s$

- 12 h irradiation maximum
  → ρa=21.50[ARTs/mm²]
- T=30nm (Hypothesis)
  (Googen and Wagner, 2000)
- Th/U=1



U concentration and age limit

U (ppm)	ART-age (10^4 year)
1.0	48.0
2.0	24.0
3.0	16.0
4.0	12.0
5.0	9.6
6.0	8.0
7.0	6.9