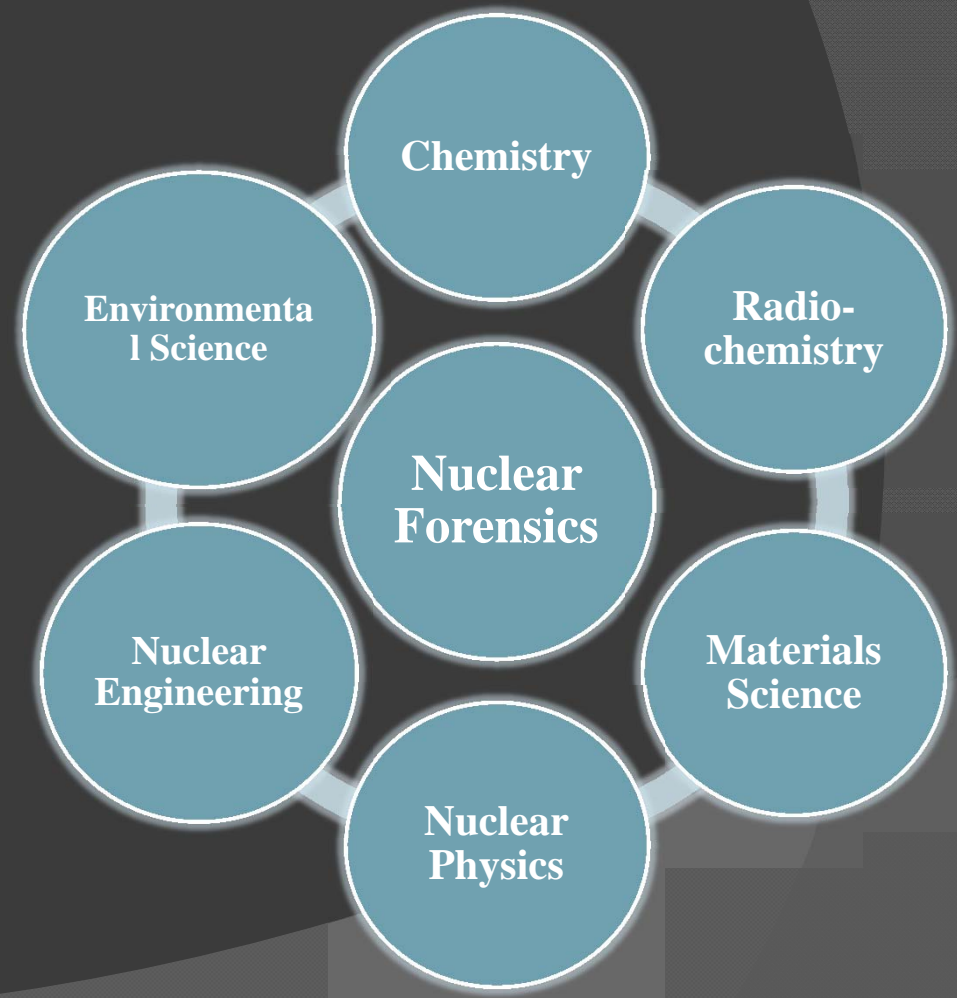


# RADIOLOGICAL CHRONOMETRY OF URANIUM METAL SAMPLES

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# Nuclear Forensics Analysis of Uranium Metal

- ◉ What is nuclear forensics?
  - the collection and analysis of nuclear or radiological material to support investigations into the trafficking or use of those materials



# Nuclear Forensics

- ⦿ Important questions to ask:
  - What type of material is the sample?
  - When was the material removed from legitimate control?
  - Where or how was the material produced?
  - **What is the provenance of the material?**
  - **What is the *age* of the sample?**
    - Age is one of several signatures

# Samples for Analysis



**U-rod**



**F-Element  
Solid**



**F-Element  
Drillings**



# U Metal Sample Preparation

- Etching procedure
- Serial rinse using 8M  $\text{HNO}_3$ , Milli-Q water, and acetone

- Dissolve in  $\text{HNO}_3$  and HF
- Dilute with Milli-Q water to obtain a 6M  $\text{HNO}_3$  solution
- Primary and secondary dilutions made with 2%  $\text{HNO}_3$

- Aliquots traced with either  $^{233}\text{U}$  or  $^{229}\text{Th}$
- Analyze U by MC-ICP-MS

- Separate and purify thorium (AG and TEVA)
- Analyze Th by MC-ICP-MS



# Radiochronometry: Age Dating

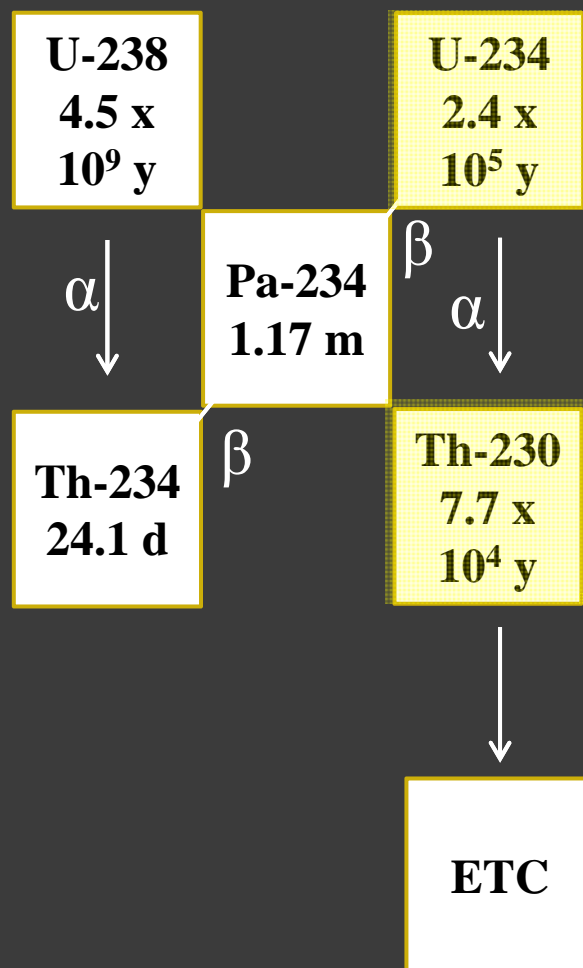
Does etching procedure affect the radiological age of uranium metal samples?

## Uranium Metal Etching Procedures

- ⦿ U-rod-1
  - 8M  $\text{HNO}_3$  + Aqua Regia followed by 8M  $\text{HNO}_3$
- ⦿ U-rod-2
  - 8M  $\text{HNO}_3$
- ⦿ U-rod-3
  - 8M  $\text{HNO}_3$
- ⦿ U-rod-4
  - 8M  $\text{HNO}_3$  + Conc.  $\text{HCl}$  x 2



# Age Determination for Uranium

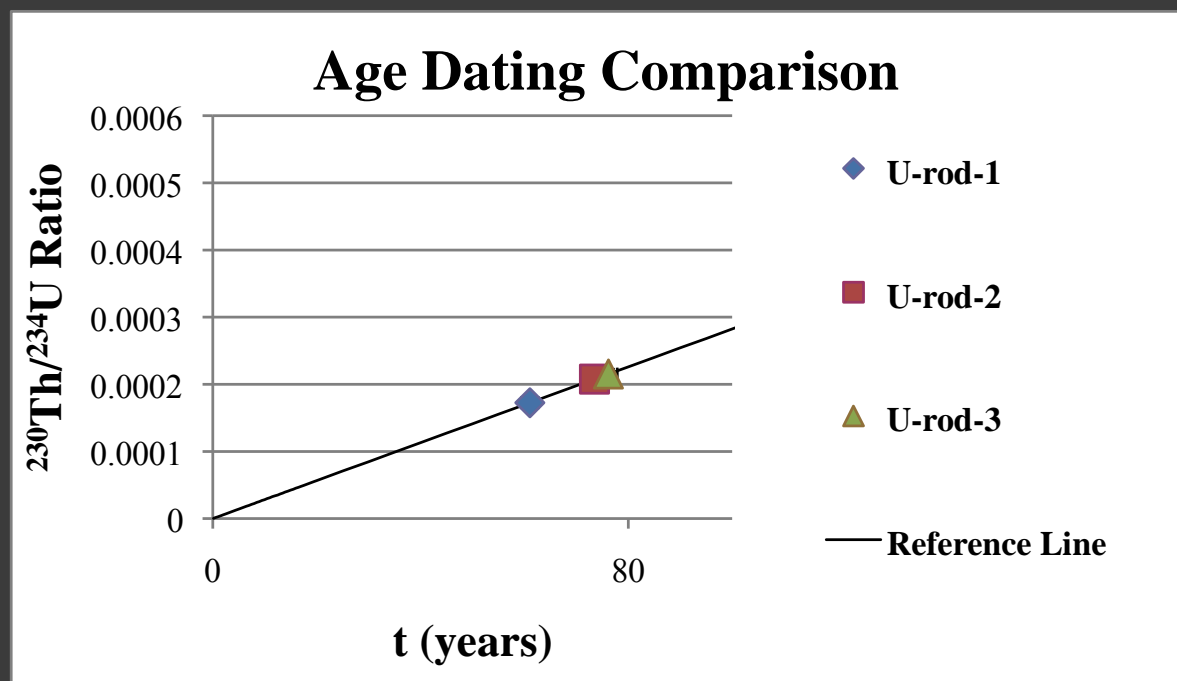


$$t = \frac{1}{(\lambda_{234} - \lambda_{230})} \times \ln \left[ 1 + \frac{N_{230Th}}{N_{234U}} \times \frac{(\lambda_{234} - \lambda_{230})}{\lambda_{234}} \right]$$

$$t_{1/2} = \frac{\lambda}{\ln(2)}$$

- $\lambda_{234}$  and  $\lambda_{230}$  are decay constants for  $^{234}\text{U}$  and  $^{230}\text{Th}$ , respectively
- $N_{230\text{Th}}$  and  $N_{234\text{U}}$  are the #'s of  $^{230}\text{Th}$  and  $^{234}\text{U}$  atoms, respectively
- $^{234}\text{U}$  half-life ( $t_{1/2}$ ) =  $245,250 \pm 490$  years ( $2\sigma$ )
- $^{230}\text{Th}$  half-life ( $t_{1/2}$ ) =  $75,690 \pm 230$  years ( $2\sigma$ )

# Age Dating Results: U-rod



Error bars are within point and represent  $3\sigma$ .

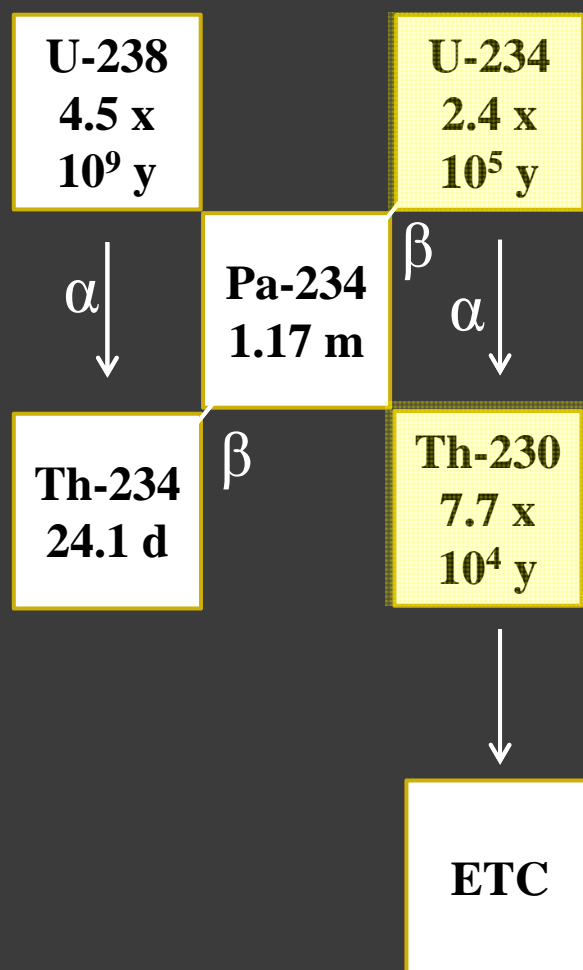
Sample description	U, g/g	Predicted Production Date
U-rod-1	$0.984 \pm 0.005$	July, 1950 $\pm 1.6$ years
U-rod-2	$0.886 \pm 0.008$	Dec., 1938 $\pm 1.7$ years
U-rod-3	$0.900 \pm 0.008$	Apr., 1936 $\pm 1.7$ years

\* U uncertainties are given as the combined standard uncertainty ( $1\sigma$ ).

\* Analysis date ( $t=0$ ) for all metal samples is March 19, 2012 besides U-rod-1, which is Aug. 1, 2011.



# Age Determination for Uranium



⦿ Uranium radiochronometry requires:

- Complete removal of  $^{230}\text{Th}$  at purification/fabrication.
- Removal of surface contamination.
- Complete separation of uranium and thorium during chemical analysis.

# Radiochronometry: Age Dating

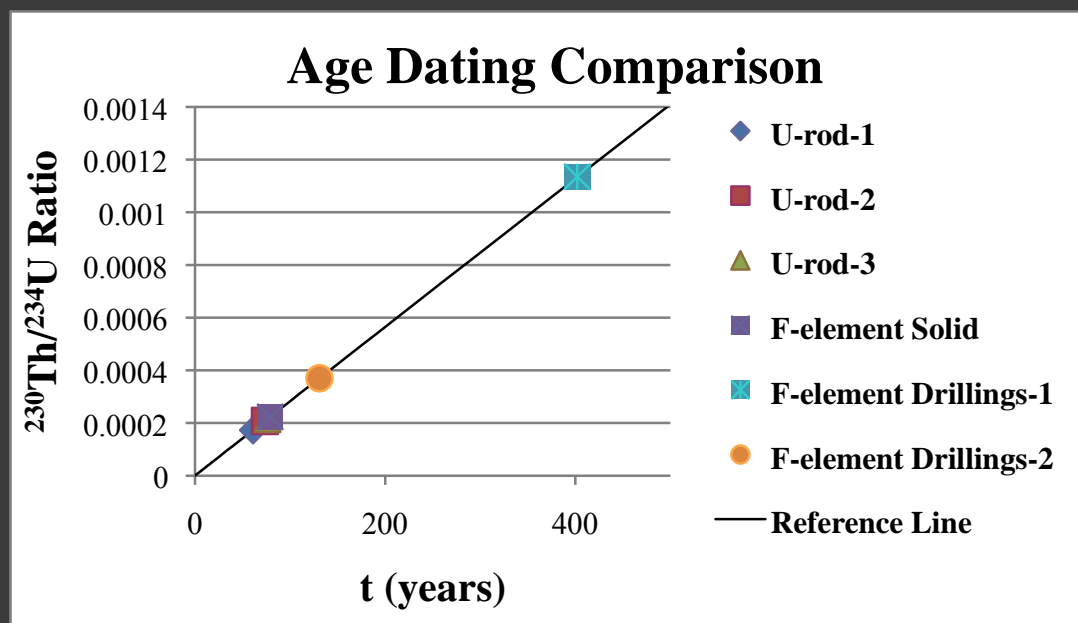
Does etching procedure affect the radiological age of uranium metal samples?

## Uranium Metal Etching Procedures

- ⦿ F-element Solid
  - 8M  $\text{HNO}_3$
- ⦿ F-element Drillings 1
  - NONE
- ⦿ F-element Drillings 2
  - NONE



# Age Dating Results: F-elements



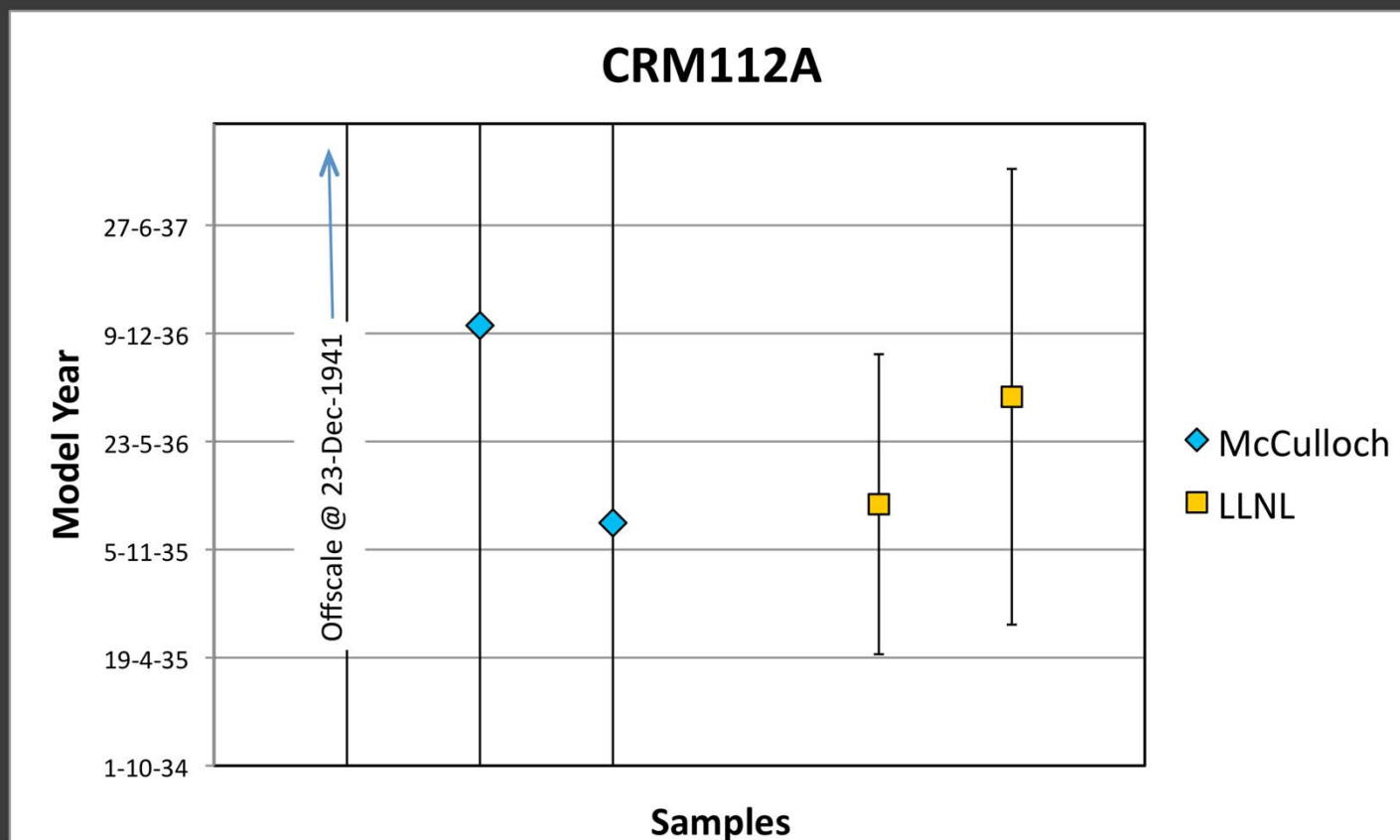
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<b>F-element Solid</b>	$1.003 \pm 0.009$	May, 1933 $\pm 1.8$ years
<b>F-element Drillings-1</b>	$0.929 \pm 0.007$	402 years old $\pm 8.1$ years
<b>F-element Drillings-2</b>	$0.932 \pm 0.008$	131 years old $\pm 2.9$ years

\* U uncertainties are given as the combined standard uncertainty ( $1\sigma$ ).

\* Analysis date ( $t=0$ ) for all metal samples is March 19, 2012 besides U-rod-1, which is Aug. 1, 2011.

# CRM 112A Age Dating



Error bars are within point and represent  $2\sigma$ .

Etching procedure for reference material according to certificate is 8M  $\text{HNO}_3$ . Standard was prepared by NBS from essential pure U in the 1950s. Thus, should have an age of ~60 years in 2010.

# Conclusions

Does etching affect radiological age of uranium metal samples?

- Radiochronometry may be affected by the method of sample preparation.
- Vigorous etching appears necessary to remove surface uranium and thorium that may differ in isotopic composition from the bulk material.
- This research demonstrates that severely weathered samples of uranium metal require vigorous surface etching to avoid introducing a systematic bias in determining radiological age.



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