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USE OF 10^{12} AND 10^{13} OHM RESISTOR AMPLIFIERS FOR URANIUM ISOTOPIC MEASUREMENTS BY TIMS AND MC- ICPMS

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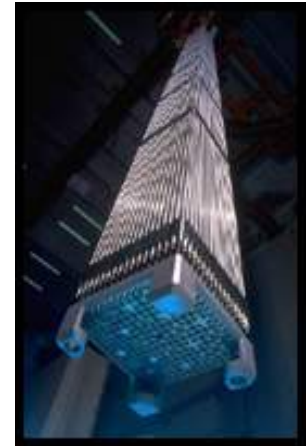
CEA Saclay, DEN / DANS / DPC / SEARS / Laboratoire de
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27 July 2016, Goldschmidt 2016 Yokohama



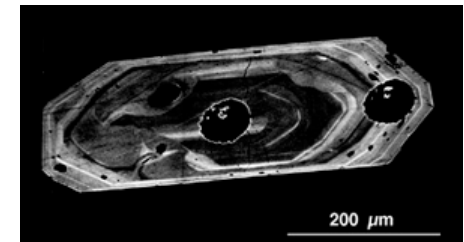
■ NUCLEAR DOMAIN

- Isotopic characterization at all steps of **nuclear fuel cycle**
- Validation of **neutronic calculation codes**
- IAEA **safeguards**
- **Nuclear forensics**. Source discrimination and datation (U-Th)
- Certification of nuclear materials (**metrology**)



■ EARTH SCIENCE

- **Datation**: U-Pb, U-Th
- **Sources discrimination**: paleoclimatology, hydrology
- Study of **isotopic fractionation** ($^{235}\text{U}/^{238}\text{U}$ fractionation)



■ Minor uranium isotope ratio measurements (^{234}U and ^{236}U)

- Isotopic ratios determined **on MC instruments** (TIMS, MC-ICPMS). Minor isotopes are generally measured **SEM** or **Daly electrode coupled to a photomultiplier**

Drawbacks: - Many settings must be performed (**linearity, dead time, gain** between SEM or Daly electrode and Faraday cup)
- Limited **time of life**

■ Advantages of 10^{12} et 10^{13} ohm amplifiers

- Increased by a factor 10 to 100 relative to 10^{11} ohm amplifier → the **signal to noise ratio** is increased by a **factor 3 to 10**
- Easy **calibration, flexibility** (choice of the amplifier resistor considering the application)
- **10^{12} ohm:** developed in 2007 (Tuttas et al.). **Applications:** K, Nd, Pb, Hf, S, W. Measured signals in the order of few mV but limited **below the mV**.
- **10^{13} ohm:** developed in 2013-2014. **Applications:** Nd, Pb, Sr. Signals lower than mV **and generally measured by SEM or Daly electrode**

■ Systematic study of certified reference materials (IRMM)

	^{234}U	^{235}U	^{236}U	^{238}U
IRMM 183	0,0019688 %	0,32049 %	0,0147858 %	99,66276 %
IRMM 184	0,0052752 %	0,72096 %	0,000012356 %	99,2738 %
IRMM 185	0,0175913 %	1,96574 %	0,00028316 %	98,01639 %
IRMM 186	0,028479 %	2,98430 %	0,0032217 %	96,98399 %
IRMM 187	0,036935 %	4,5167 %	0,0068683 %	95,4395 %

- **TIMS** (Triton Plus) and **MC-IPMS** (Neptune Plus) measurements
- **Choice of the ohm resistor** considering the atomic abundance of ^{234}U and ^{236}U
- **Internal normalisation** $^{235}\text{U}/^{238}\text{U}$ to better compare the **performances** of 10^{12} and 10^{13} ohm resistors for **low signals**
- **Gain calibration** of 10^{13} relative to 10^{11} ohm resistors performed using an **Nd isotopic standard** → **reproducibility** around 40 ppm

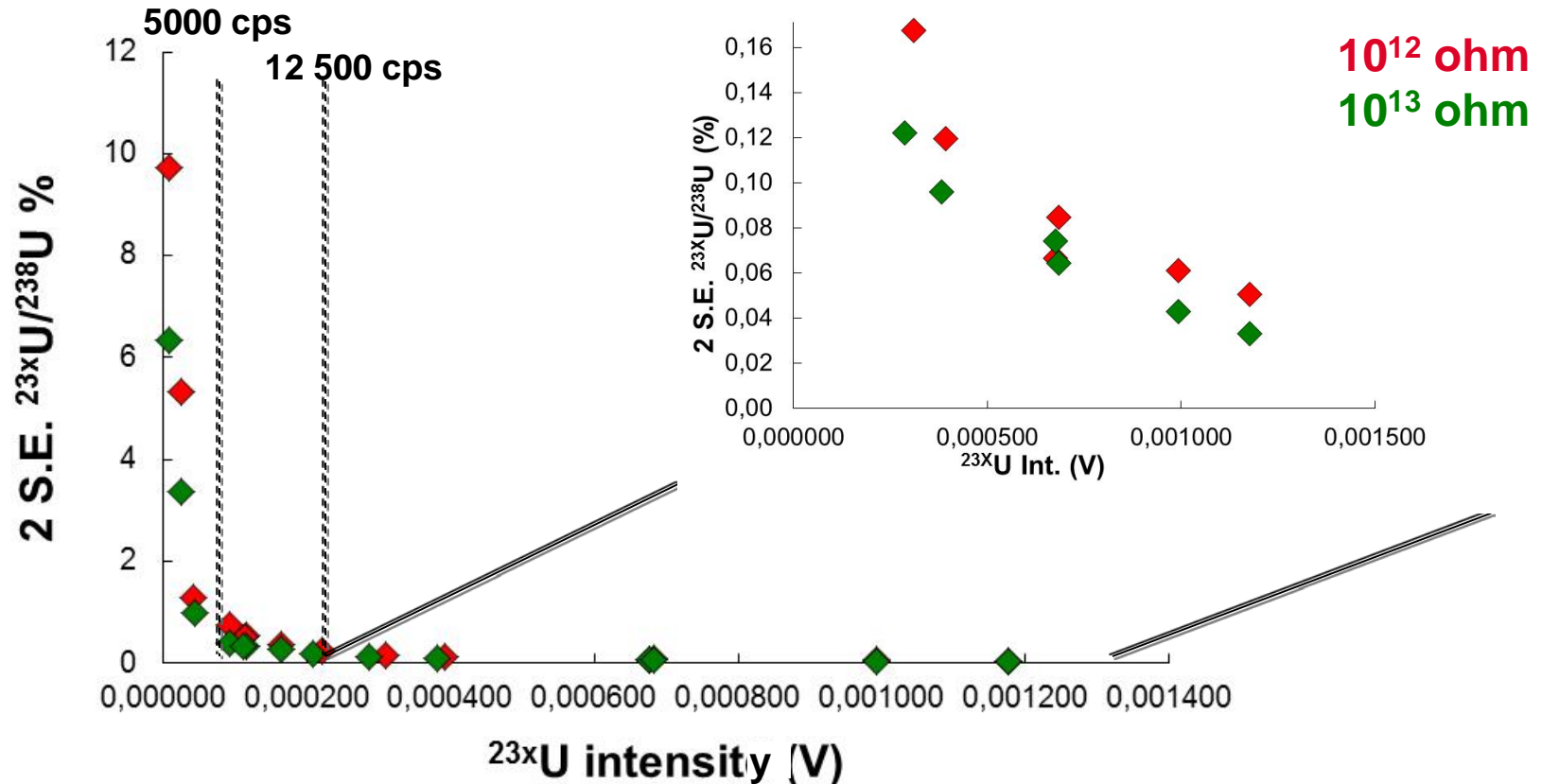
■ Details of measurement procedure by static multicollection

	L2	L1	Ax	H1	H2
	10 ¹² ou 10 ¹³	10 ¹¹	10 ¹² ou 10 ¹³	10 ¹¹	10 ¹¹
Seq. 1	²³⁴ U	²³⁵ U	²³⁶ U	237	²³⁸ U
Seq. 2	233.5	234.5	235.5	236.5	237.5
Seq. 3	234.5	235.5	236.5	237.5	238.5



- ²³⁸U signal between 3 and 4 volts. Number of **measurement cycles = 80** (integration time = 16.777 secondes).
- **Abundance sensitivity corrections:** measurements of half masses (233.5 and 234.5 for 234 mass and 235.5 and 236.5 for 236 mass measured on the same ohm resistors)

- **Internal** reproducibility obtained according to the ion beam intensity



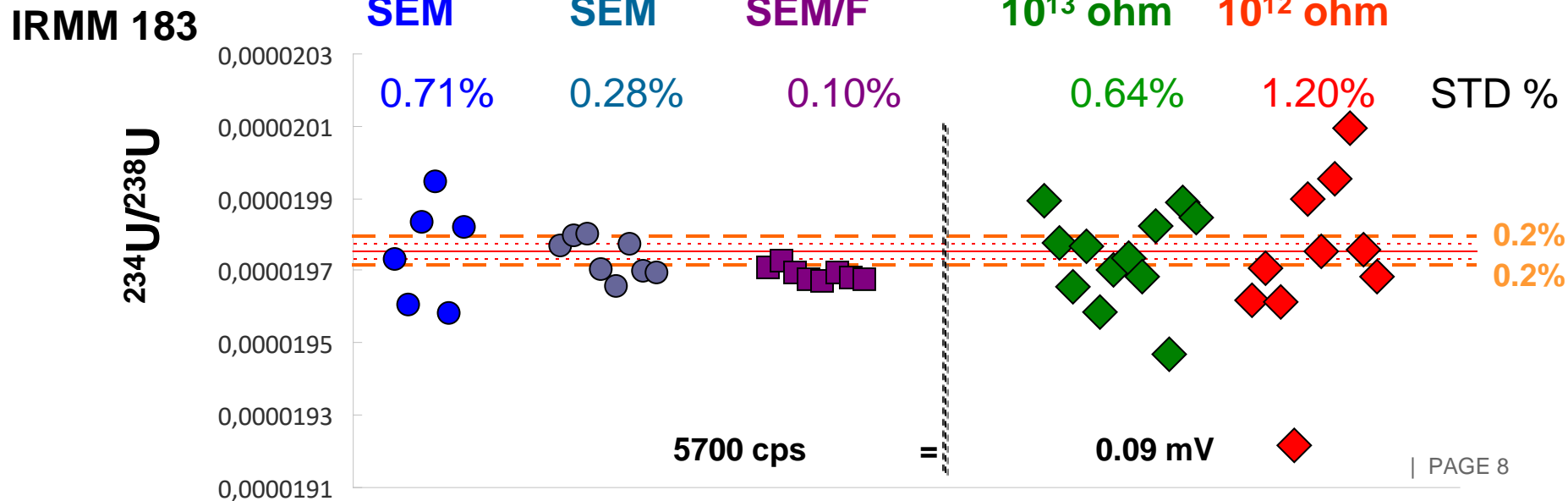
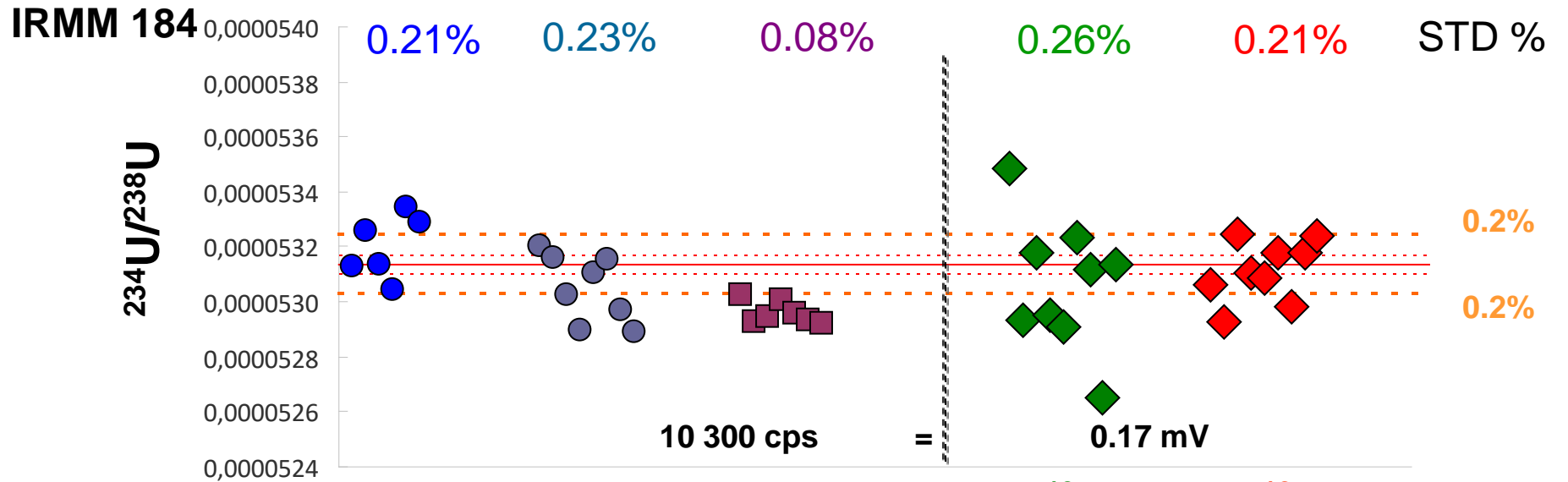
- **Signals between 0.08 and 0.2 mV (5000 to 12500 on SEM)** Comparison of measurements 10¹², 10¹³ ohm and SEM → IRMM 183 et 184

- Measurement procedure by dynamic multicollection with axial SEM (+ electrostatic filter)

	L2	L1	Ax (SEM)	H1	H2	H3
Seq. 1			^{234}U			^{238}U
Seq. 2			^{235}U		^{238}U	
Seq. 3	233.5	234.5	235.5			
Seq. 4	^{234}U	^{235}U	^{236}U	^{238}U		
Seq. 5	234.5	235.5	236.5			

- ^{238}U signal between 3 and 4 volts. Number of **measurement cycles = 40** (integration time = 16.777 secondes for ^{234}U and ^{236}U and 8.839 secondes for ^{235}U and ^{238}U)
- **Abundance sensitivity corrections:** measurements of half masses (235.5 and 236.5 for 236 mass measured on SEM)
- **Internal correction of Gain F/SEM gain:** ^{235}U measured on SEM (Seq. 2) and Faraday (Seq. 4)

TIMS: COMPARISON 10^{12} , 10^{13} , SEM (5000 à 12 500 cps)



■ Measurement procedure by multicollection

	L2	L1	Ax	H1	H2
	10^{12}	10^{11}	10^{11}	10^{11}	10^{11}
Seq. 1	^{234}U	^{235}U	^{236}U	237	^{238}U
Seq. 2	233.5	234.5	235.5	236.5	237.5
Seq. 3	234.5	235.5	236.5	237.5	238.5



- ^{238}U signals between 4 and 45 volts. Number of **measurement cycles = 30** (integration time = 8.4 secondes)
- **Abundance sensibility corrections**: measurement of half masses (233.5 and 234.5 for 234 mass and 235.5 and 236.5 for 236 mass measured on the same ohm resistors)
- **Measurements of the certified isotopic standard IRMM 184** at different signals measured on the ^{234}U mass on 10^{12} ohm resistor amplifier

■ External reproducibility and accuracy on the IRMM 184 standard

IRMM 184

0.82%

0.41%

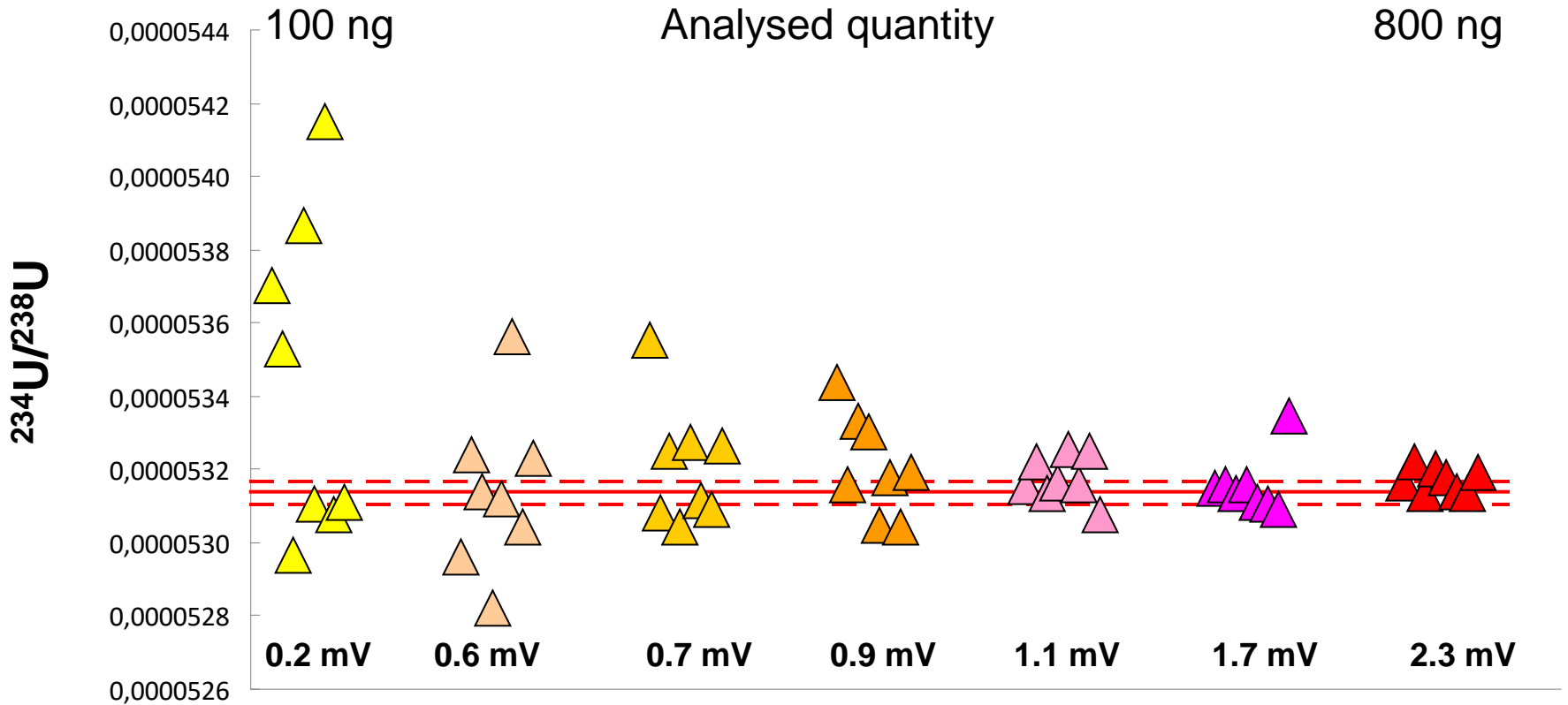
0.34%

0.26%

0.11%

0.15%

0.06%



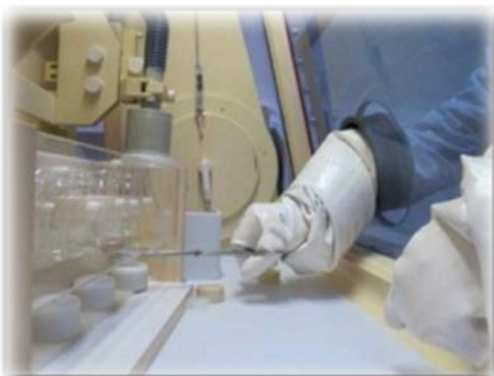
■ Measurements of **234, 235 et 238** on Faraday cups

■ Method **validation** : accuracy < 0,1% (signal of 2.3mV signal on ²³⁴U)

- **New analytical procedures for the determination of uranium isotope ratios by TIMS with measurements of 234 and 236 uranium isotopes on $10^{12} \Omega$ et $10^{13} \Omega$ resistors**
 - **Flexibility**
 - **Robustness** in comparison to SEM measurements
 - **Reproducibility** better than those obtained by SEM for signals $> 12\,500$ cps ($=0.2$ mV)

- **MC-ICPMS measurements and measurements of 234 isotope on $10^{12} \Omega$**
 - Necessity to take into account of **baseline corrections and abundance sensibility corrections**
 - **Reproducibility** obtained lower than 0,1% for signals in the order of few mV

- **Perspectives**
 - **MC-ICPMS** measurements of uranium minor isotopes on **$10^{13} \Omega$**
 - Development of **coupling between separative techniques and MC-ICPMS MC** with measurements of uranium minor isotopes on **$10^{12} \Omega$**



Thanks for your attention



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