



HAL
open science

Use of 10^{12} and 10^{13} ohm resistor amplifiers for uranium isotopic measurements by TIMS and MC-ICPMS

H. Isnard, M. Aubert, A. Nonell, F. Chartier

► To cite this version:

H. Isnard, M. Aubert, A. Nonell, F. Chartier. Use of 10^{12} and 10^{13} ohm resistor amplifiers for uranium isotopic measurements by TIMS and MC-ICPMS. Goldschmidt 2016 - International conference on geochemistry and related subjects, Jun 2016, Yokohama, Japan. cea-02439460

HAL Id: cea-02439460

<https://cea.hal.science/cea-02439460>

Submitted on 26 Feb 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

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

ISNARD H., AUBERT M., NONELL A., CHARTIER F;

CEA Saclay, DEN / DANS / DPC / SEARS / Laboratoire de
développement Analytique Nucléaire Isotopique et Élémentaire

27 July 2016, Goldschmidt 2016 Yokohama



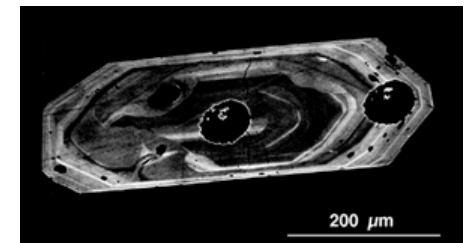
■ NUCLEAR DOMAIN

- 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

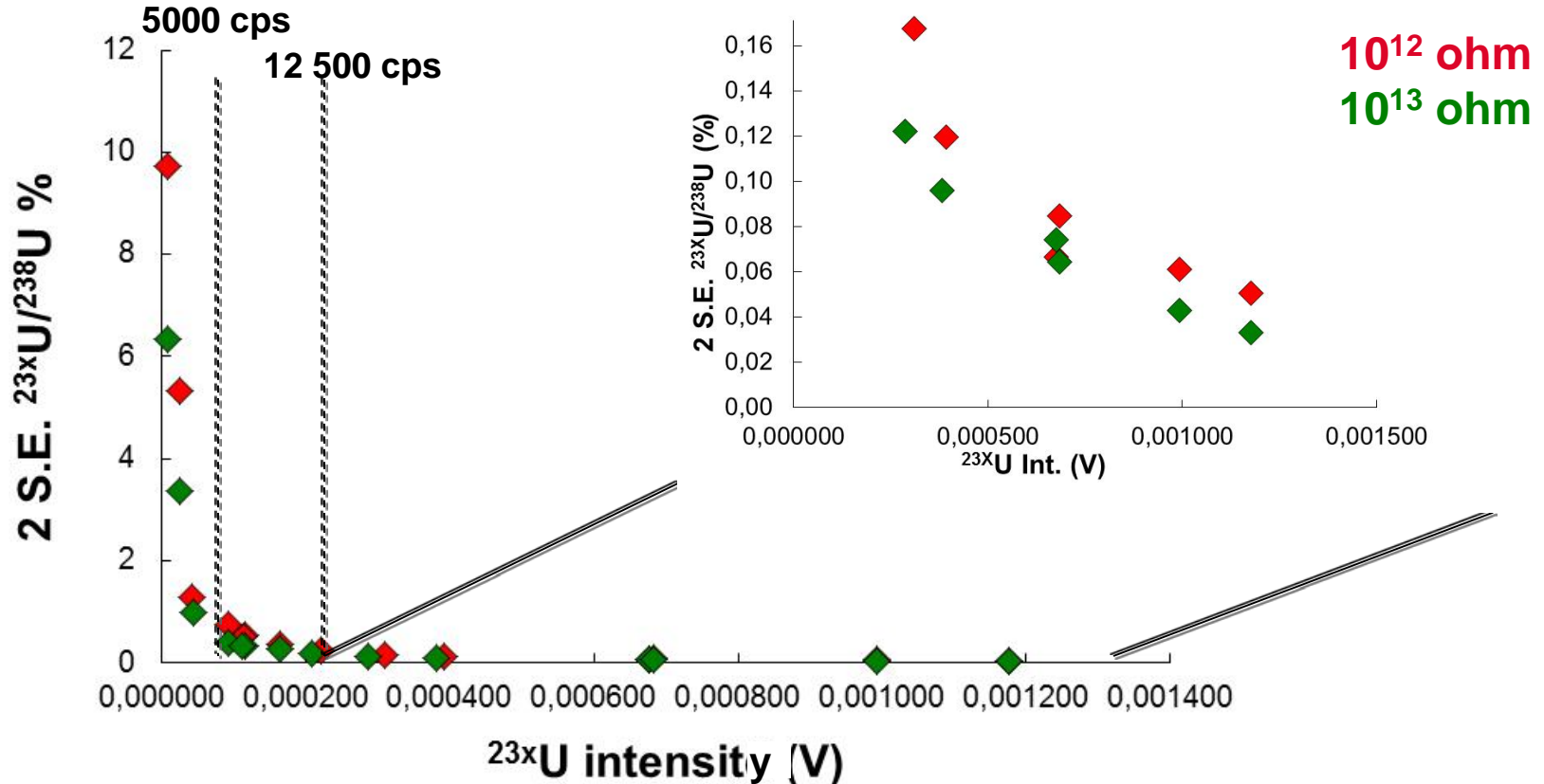
■ Details of measurement procedure by static multicollection

	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



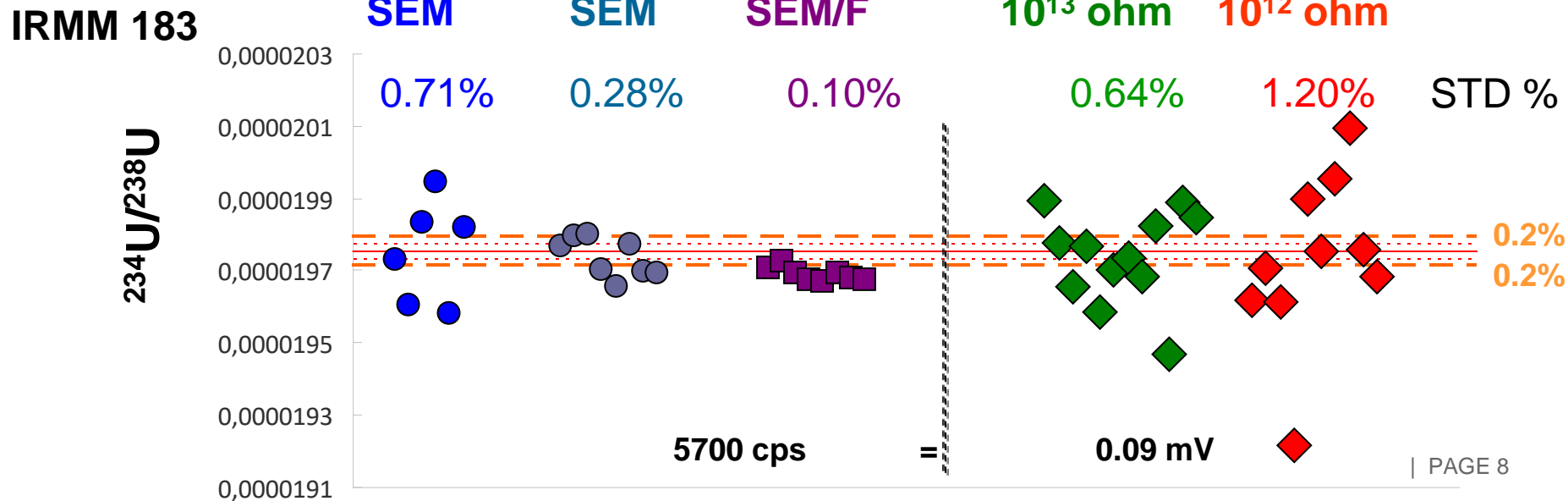
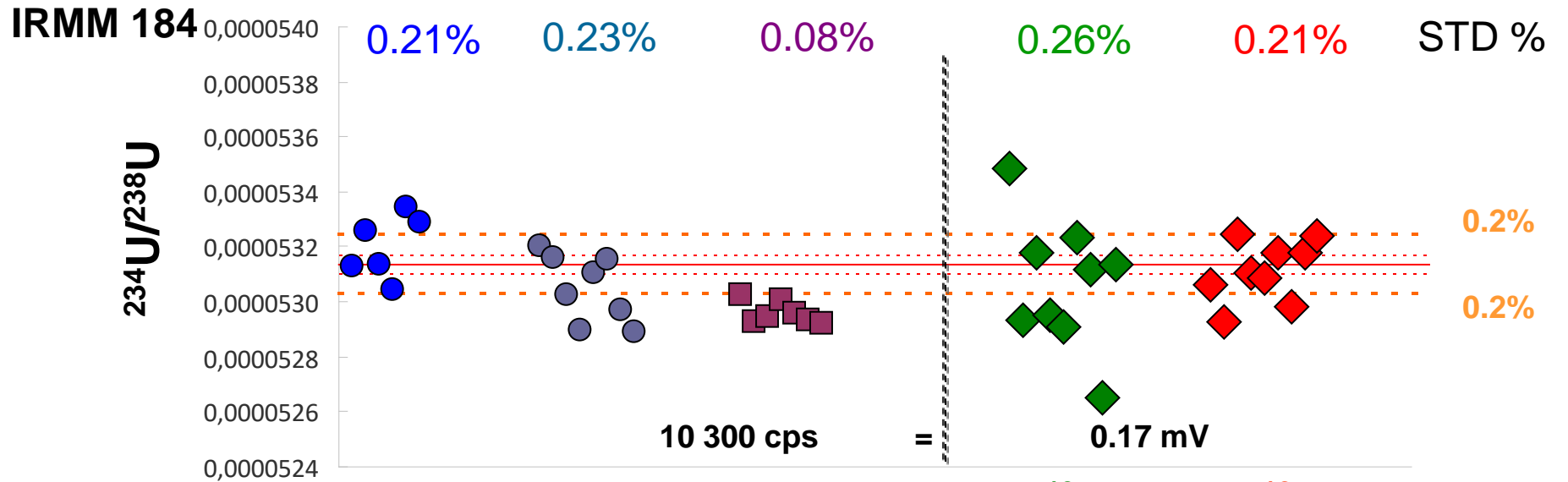
- Signals between 0.08 and 0.2 mV (5000 to 12500 on SEM) Comparison of measurements 10¹², 10¹³ ohm and SEM → IRMM 183 et 184

- Measurement procedure by dynamic multicollecion 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%

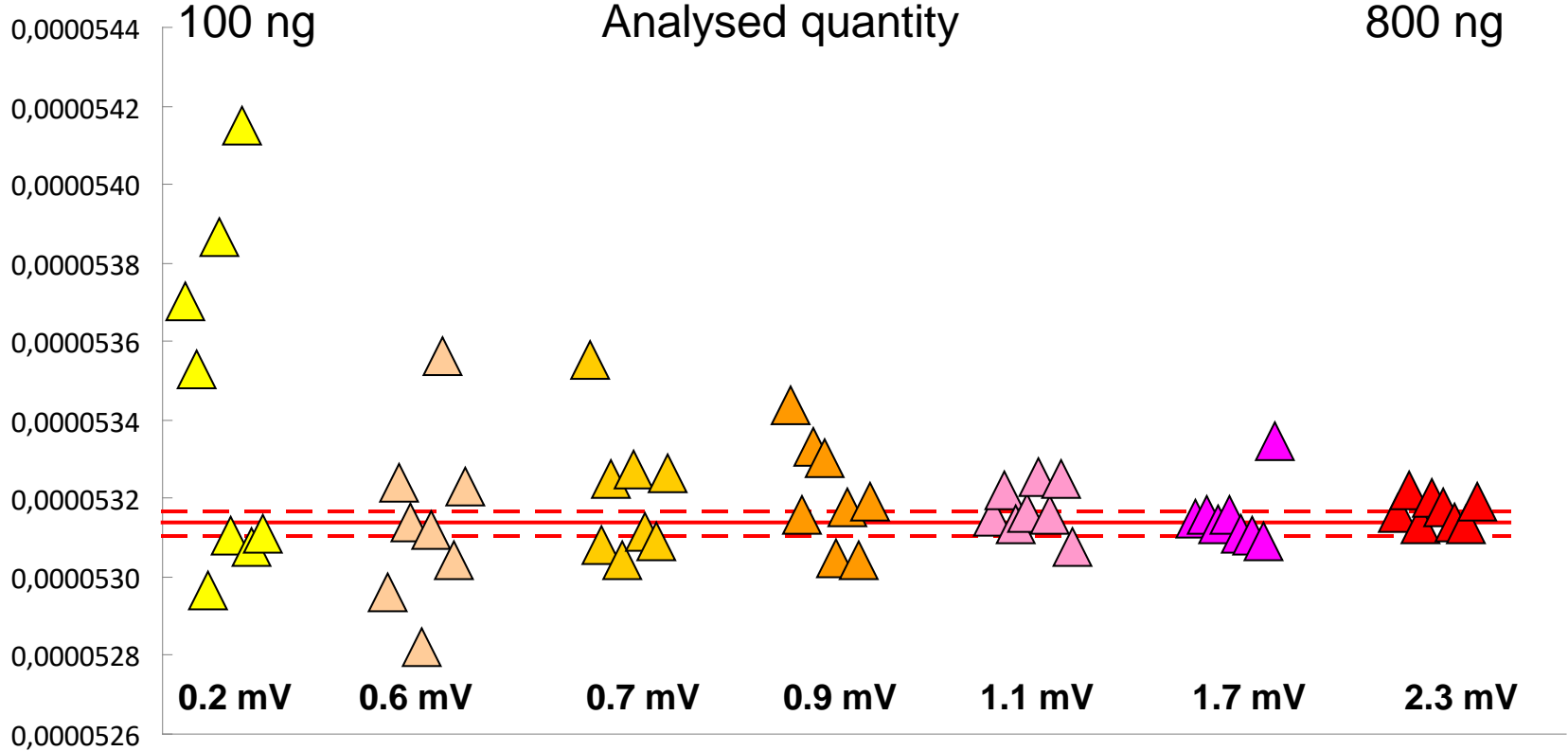
0.06%

100 ng

Analysed quantity

800 ng

$^{234}\text{U}/^{238}\text{U}$



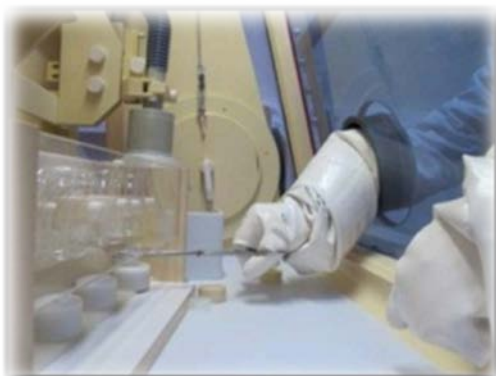
■ Measurements of **234, 235 et 238** on Faraday cups

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

- **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



Commissariat à l'énergie atomique et aux énergies alternatives
Centre de Saclay | 91191 Gif-sur-Yvette Cedex
T. +33 (0)1 69 08 80 79 | F. +33 (0)1 69 08 54 11

Etablissement public à caractère industriel et commercial | R.C.S Paris B 775 685 019

DEN
DPC
SEARS
LANIE