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Measurements of long-lived ^{121m}Sn and ^{126}Sn nuclides in Low and Intermediate Level Nuclear Waste

Céline Gautier, J.-P. Degros, M. Coppo, P. Fichet, F. Goutelard, R. Brennetot

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C. Gautier¹, J.P. Degros¹, M. Coppo¹, P. Fichet¹, F. Goutelard² and R. Brennetot¹

¹Operator Support Analyses Laboratory, Atomic Energy Commission, CEA Saclay, DEN/DANS/DPC/SEARS/LASE, Building 459, PC171, 91191 Gif-sur-Yvette Cedex, FRANCE
celine.gautier@cea.fr

²CEA/DEN/MAR/DEIM/Nuclab, Atomic Energy Commission, CEA Marcoule, Building 109, BP 17 171, 30207 Bagnols-sur-Geze, FRANCE

Context

In France, nuclear waste are managed by the National Radioactive Waste Management Agency (ANDRA). Several repository sites have been built in order to accommodate nuclear waste packages. One is dedicated to waste containing short-lived radionuclides with radioactivity at Low and Intermediate Level. Criteria for 143 radionuclides have been defined by ANDRA which guarantees the safety of the facility. Among this long list, Sn-121m and Sn-126 have to be declared as soon as their activities are over 10E-3 Bq/g [1]. Both tin isotopes are fission products obtained according to thermal fission yields of 0.00003 % and 0.06 %, respectively. Sn-121m can also be produced by activation of tin present as an additive in zircalloys and as an impurity in inconels and steels used in the nuclear industry. Sn-121m decays with a half-life of 55 years whereas Sn-126 is a very long-lived radionuclide with a half-life estimated at around 10E5 years. Because of the long half-life and the unknown impact of high energy gamma emissions of Sn-126, ANDRA allows a very low Sn-126 activity limit (2.7 Bq/g) in waste packages. Due to their potential low activities in nuclear wastes, both tin nuclides need to be separated from the matrices and concentrated through chemical separations prior to any measurement.

Radiochemical protocol

⇒ Q ICPMS (Elan DRCe, Perkin Elmer):

- Measurement of ¹²⁶Sn activity



$$A(^{126}\text{Sn}) = \frac{N \times \ln 2 \times [^{126}\text{Sn}]_{\text{ICPMS}}}{M \times T_{196\,000 \text{ years}} [2]}$$

Digestion after addition of a few µg of natural tin carrier and ¹¹³Sn spiker

Co-precipitation with Al(OH)₃ / Dissolution in 8M HCl

Preliminary separation on anion exchange AG1X8 resin

Co-precipitation with MoS₃ / Dissolution in 69% HNO₃ + HCl

Selective step

Ethylation in acetic acid buffer ⇒ SnEt₄

Purification of the organic phase on Silicagel column

Back-extraction of Sn via degradation of tetra-ethyl Sn

Co-precipitation with Al(OH)₃ / Dissolution in 6M HCl

Final purification on anion exchange AG1X4 resin with Ultrex reagents (0.5M HNO₃ + 0.01M HF)

Purification/separation steps

⇒ Gamma spectrometer (Ge detector, Ortec GMX-15185):

- Measurement of ^{121m}Sn activity
- Measurement of ¹¹³Sn activity for the determination of the radiochemical separation yield



⇒ Radiochemical yield: 30% up to 70% depending on the matrix

Q ICPMS
¹²⁶Sn

γ Spectrometry
¹¹³Sn (392 keV)

γ Spectrometry
^{121m}Sn (37 keV)

Validation

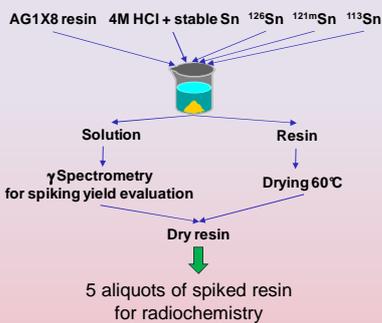
In-house tin standards preparation

No availability of commercial ^{121m}Sn and ¹²⁶Sn sources

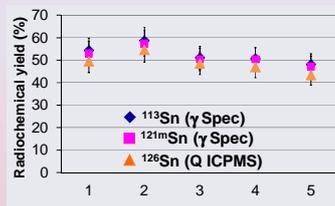
Application to radioactive zircalloys ⇒ in-house ^{121m}Sn source

Application to spent nuclear fuel dissolution solutions ⇒ in-house ¹²⁶Sn source

Spiking of ion exchange resins with ¹¹³Sn*, ^{121m}Sn, ¹²⁶Sn sources (*CERCA commercial source)



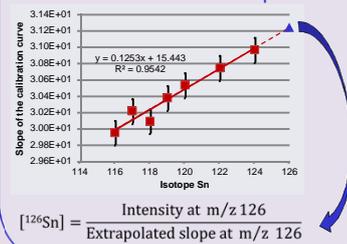
Results of 5 radiochemical replicates



⇒ Same radiochemical yields whatever the studied tin isotopes

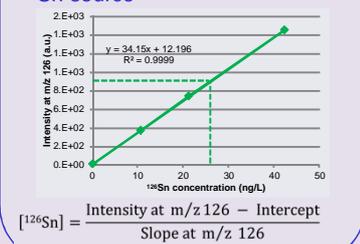
Determination of ¹²⁶Sn activity

❖ Method 1: Extrapolation of the slope of the ¹²⁶Sn calibration curve from other natural tin isotopes



$$[^{126}\text{Sn}] = \frac{\text{Intensity at } m/z \text{ 126}}{\text{Extrapolated slope at } m/z \text{ 126}}$$

❖ Method 2: ¹²⁶Sn calibration curve obtained from the in-house ¹²⁶Sn source



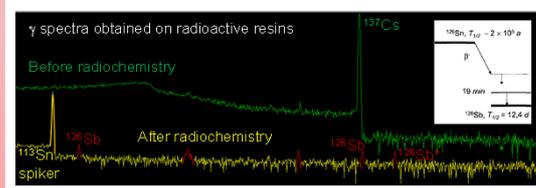
$$[^{126}\text{Sn}] = \frac{\text{Intensity at } m/z \text{ 126} - \text{Intercept}}{\text{Slope at } m/z \text{ 126}}$$

	[¹²⁶ Sn] (ng/L) obtained from Method 1	[¹²⁶ Sn] (ng/L) obtained from Method 2	Difference (%)
Sample 1	36.2	36.7	-1.4
Sample 2	12.3	11.7	4.8

⇒ No significant difference between the two methods

Applications

Real matrices: radioactive steels and ion-exchange resins



	^{121m} Sn activity (Bq.g ⁻¹)
Steels	50 → 1000



	¹²⁶ Sn activity (Bq.g ⁻¹)
Resins	0.1 → 10

Conclusions

A radiochemical protocol was developed on synthetic samples to separate tin from various matrices and concentrate it prior to any nuclear or mass spectrometry measurements. As no ^{121m}Sn or ¹²⁶Sn sources are commercially available, the protocol was first applied to zircalloys and spent nuclear fuel dissolution solutions to obtain in-house standards. The radiochemical procedure was validated subsequently on ion exchange resins spiked with ¹¹³Sn, ^{121m}Sn and ¹²⁶Sn. The developed radiochemical procedure enables to measure ^{121m}Sn and ¹²⁶Sn in Low and Intermediate Level Nuclear Waste with detection limits of 20 Bq/g and 0.02 Bq/g respectively.

[1] ANDRA, Spécifications d'acceptation des colis de déchets radioactifs au centre de l'Aube - Spécification technique générale, ACO.SP.ASRE.99.001 indice D, 2014.
[2] P. Bienvenu et al., Determination of ¹²⁶Sn half-life from ICP-MS and gamma spectrometry measurements, Radiochim. Acta 97, 687-694 (2009)