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| 12 13 | Accurate determination of Curium and Californium isotopic ratios by |
| 14 | inductively coupled plasma quadrupole mass spectrometry (ICP-QMS) in |
| 15 | ²⁴⁸ Cm samples for transmutation studies |
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46 Abstract

47 The French Atomic Energy Commission has carried out several experiments including the 48 mini-INCA (INcineration of Actinides) project for the study of minor-actinide transmutation 49 processes in high intensity thermal neutron fluxes, in view of proposing solutions to reduce the radiotoxicity of long-lived nuclear wastes. In this context, a Cm sample enriched in ²⁴⁸Cm 50 51 (~97 %) was irradiated in thermal neutron flux at the High Flux Reactor (HFR) of the Laue-52 Langevin Institute (ILL). This work describes a quadrupole ICP-MS (ICP-QMS) analytical 53 procedure for precise and accurate isotopic composition determination of Cm before sample 54 irradiation and of Cm and Cf after sample irradiation. The factors that affect the accuracy and 55 reproducibility of isotopic ratio measurements by ICP-QMS, such as peak centre correction, 56 detector dead time, mass bias, abundance sensitivity and hydrides formation, instrumental 57 background, and memory blank were carefully evaluated and corrected. Uncertainties of the 58 isotopic ratios, taking into account internal precision of isotope ratio measurements, peak 59 tailing, and hydrides formations ranged from 0.3% to 1.3%. This uncertainties range is quite 60 acceptable for the nuclear data to be used in transmutation studies.

61

62 **1. Introduction**

63 Management of nuclear waste produced during the burn-up of nuclear fuel is one of the 64 crucial challenges in this century. A very promising way under investigation for the 65 management of radioactive waste is the transmutation of long-lived radionuclides in a fast 66 neutron nuclear reactor into shorter lived or stable ones [1-3]. Therefore, transmutation of 67 long-lived nuclides has been investigated in various systems for characterisation of their 68 nuclear data. Due to their limited role in conventional fuel cycles, minor actinides (MA) have 69 not been extensively studied and their nuclear parameters are not always known with the 70 precision required for transmutation dedicated system [4, 5]. In this context, the French 71 Atomic Energy Commission has carried out several experiments including the mini-INCA 72 (INcineration of Actinides) project. The objective of this project is to extend the field of 73 investigation of minor actinides (1), which play an important role in the nuclear waste 74 inventories for future nuclear energy scenarios, and (2) for which nuclear parameters are 75 determined with significant discrepancies between different nuclear data libraries. Several 76 radionuclides have been previously examined [6, 7].

Part of these studies is based on experimental results obtained after irradiation of pure isotopes in the High Flux Reactor (HFR) of the Laue-Langevin Institut (ILL). The isotopic composition measurements of the studied isotopes before and after irradiation by mass 80 spectrometric techniques are used to improve or complete nuclear data libraries. As part of this program, a Cm sample enriched in 248 Cm (~97 %) was irradiated in thermal neutron flux 81 in the HFR of the ILL. The main objective of the program is to determine ²⁴⁸Cm neutron 82 83 capture cross-sections and to study part of the isotopic chains of Cm, Bk, and Cf. The simplified evolution chain of ²⁴⁸Cm under neutron irradiation is presented in figure 1. Figure 84 1 shows that the short-lived ²⁴⁹Cm (64.15 min) generated by neutron capture of ²⁴⁸Cm, decays 85 into ²⁴⁹Bk (330 d), which in turn decays to ²⁴⁹Cf (351 y). Isotopes of ²⁵⁰Cf (13 y), ²⁵¹Cf (898 86 y), and ²⁵²Cf (2.6 y) are generated by neutron capture of ²⁴⁹Cf, ²⁵⁰Cf, and ²⁵¹Cf, respectively. 87 Thermal Ionisation Mass Spectrometry (TIMS) is the technique of choice for actinides 88 89 measurements in nuclear sample when high accuracy is needed [8-10]. This method advanced analytical capacities with per-mil-level precision [11, 12] and the latest generation of these 90 91 instruments allows isotope measurements with precision of several ppm level [13, 14].



Figure 1. Simplified evolution chain of Cm-248 under neutron irradiation. After 1-year cooling period. Cm-249
 has totally decayed.

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In recent years, inductively coupled plasma source mass spectrometers equipped with multi collectors (MC-ICPMS) can attain isotope ratio precision $\leq 0.01\%$ [15-17].

97 At present, it is well documented that Inductively Coupled Plasma Quadrupole Mass 98 Spectrometry (ICP-QMS) cannot achieve the precision and accuracy in isotope ratio 99 measurements attainable in TIMS but has also been widely used in isotope ratio 100 measurements [17, 18]. Indeed, depending on the final objective of the measurement, the ICP-101 QMS accuracy might remain significantly below other sources of uncertainty (e.g. neutron 102 fluence during irradiation). Even if precision attained with this instrument (Relative Standard 103 Deviation ~ 0.1 -1%) remains inferior compared to TIMS and MC-ICPMS, simple and low 104 cost instrumentation, and simplified measurement process make ICP-QMS more practical to 105 use than TIMS or MC-ICPMS. Two basic reasons for the inferior precision of ICP-QMS 106 compared to TIMS and MC-ICPMS are the sequential rather than simultaneous isotope 107 measurement and the Gaussian peaks shape characteristic of these instruments [19]. Some 108 recent studies have been conducted in order to improve ICP-QMS performance when

- 109 measuring isotope ratios [20-22]. The precision attained with ICP-QMS is quite sufficient for 110 nuclear data relevent to this case study.
- 111 In this work, a methodology for a precise and accurate determination of isotopic composition
- of Cm sample enriched in 248 Cm (~97%), before and after irradiation in the HFR of the ILL
- 113 by ICP-QMS has been developed.

The factors that affect the accuracy and reproducibility of isotopic ratios by ICP-QMS, such as the peak centre correction, detector dead time, mass bias, abundance sensitivity and hydrides formation, instrumental background, and memory blank were carefully evaluated and corrected. Isotopic results for Cm before irradiation and Cm, Bk, and Cf after sample irradiation are presented with the full experimental and analysis procedures that were followed to obtain these data.

120

121 **2. Experimental**

122 2.1. Instrumentation settings

123 An ICP-QMS X series mass spectrometer (Thermo Electron, Winsford, UK) housed at the 124 Isotopic Elementary and Nuclear Analysis Laboratory (LANIE) of the French Atomic Energy Commission (CEA Saclay) was used in this study. The ICP-QMS, the first X series modified 125 126 with ion source and placed in a glove box to work with radioactive samples, was previously 127 described [23]. Sample introduction into the plasma was made via a quartz peltier-cooled 128 impact bead spray chamber and a quartz concentric nebulizer (1.0 mL/min). Self-aspiration 129 mode was used for the sample introduction in order to minimize discrete noise components 130 caused by rotation of the peristaltic pump.

- All of the experimental parameters were daily optimized with a multi-elements test solution in 131 order to obtain the maximum counting rates on ¹¹⁵In and ²³⁸U for this tuning. After a 132 133 verification test of key parameters (sensitivity, stability, and oxide level test), the instrument 134 is ready to perform measurements. The typical sensitivity and stability (Relative Standard Deviation) values measured in 10 minutes for both ¹¹⁵In and ²³⁸U are $\sim 4 \times 10^5$ cps/ppb and 135 <3%, respectively. Oxide level test is performed by measuring the ¹⁴⁰CeO^{+/140}Ce ratio with 136 137 typical values varying between 3 and 4%. The ICP-QMS operating and measurement 138 conditions are summarized in Table 1.
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Table 1. Instrument settings for the ICP-QMS.

| | Value/description |
|----------------------------|-------------------|
| Sample introduction system | |
| Impact bead spray chamber | |
| Concentric nebulizer | 1.0 mL/min |
| Self-aspiration mode | |
| Spray chamber temperature | 3°C |
| ICP-QMS X conditions | |
| RF power | 1400 W |
| Nebulizer gas flow | 0.8–0.9 L/min |
| Auxiliary gas flow | 0.9 L/min |
| Cool gas flow | 15 L/min |
| Data acquisition | |
| Peak width (10%) | 0.82 a.m.u. |
| Detection mode | Pulse counting |
| Scan mode | Peak jumping |

144

145 2.2 Sample preparation

Two powder samples, 11 µg and 37 µg of ²⁴⁸Cm in the form of Cm-nitrate were provided by 146 the Lawrence Berkeley National Laboratory. The samples were canned into quartz tubes 147 148 sealed under low pressure atmosphere. The first sample was sent directly to the LANIE 149 laboratory to determine Cm isotopic composition before irradiation. The second sample was 150 placed inside an aluminium container andwas irradiated for 16 days (December 3-19.2007) 151 inside the V4 irradiation channel [24] of the HFR of ILL in Grenoble-France. After irradiation 152 and a cooling period of about 1 year the capsule was sent to the LANIE laboratory. The 153 powder was dissolved with 4M HNO₃ (Normatom Prolabo). After complete dissolution, the 154 sample was diluted for isotopic analysis. All dilutions were performed in 0.5M nitric sub-155 boiling acid. This acid was obtained by dilution of sub-boiling 14M nitric acid, using an 156 EVAPOCLEAN system (Analab, France), in deionised water (resistivity 18.2 M Ω .cm) 157 obtained from a Milli Q system (Millipore, Milford, MA, USA). All the reagents were 158 prepared in a glove-box.

Non-irradiated and irradiated samples survey runs were performed in a mass range between m/z=229 and 236, in order to indentify impurities that can create oxides isobaric interferences within the mass rage of interest. For both samples, no impurities were detected.

For mass bias correction, an in-house 248 Cm/ 246 Cm = 8.937±0.018 standard solution was used. The in-house standard nitric solution was certified using TIMS by 6 independent measurements and by the total evaporation method [25, 26]. This method, takes into account the integration of all ions formed during analysis and is considered accurate because the effects of isotope fractionation that occur during the evaporation process are virtually eliminated in the final results.

168

169 **3. Results and discussion**

- 170 3.1. Analytical method
- 171 Mass spectra of non-irradiated and irradiated Cm samples present obviously a very high 172 isotopic abundance of ²⁴⁸Cm comparing to the other isotopes (Fig. 2). The electron multiplier 173 detector employed in this study could use an analog mode (ion beam $>2\times10^6$ cps) or a pulse
- 174 counting mode (ion beam $< 2 \times 10^6$ cps) for ion signal measurements.







Figure 2. Mass spectrum of non-irradiated (a), and irradiated (b) Cm samples.

178 In order to avoid high count rates on the electron multiplier and assure pulse counting as 179 detection mode for more robust conditions, measurement of Cm samples isotopic composition was not performed in one single peak jumping routine. Thus ²⁴⁸Cm/²⁴⁶Cm isotopic ratio was 180 measured first in a low concentrated solution (~4 ng/g of ²⁴⁸Cm), and in a second run the 181 ²⁴⁵Cm/²⁴⁶Cm, ²⁴⁷Cm/²⁴⁶Cm, ²⁴⁹(Bk+Cf)/²⁵¹Cf, ²⁵⁰Cf/²⁵¹Cf, ²⁵²Cf/²⁵¹Cf isotopic ratios were 182 measured in a high concentrated solution (~100 ng/g of ²⁴⁸Cm). Each measurement consisted 183 184 of twenty five consecutive integrations (cycles). Isotopes dwell time ranged from 10 ms for 185 the high abundance isotopes to 40 ms for the low abundance isotopes [27].

For a number of 100 sweeps per cycle, the total measurement times were about 3 and 10 min for the low and high concentrated solutions, respectively. The isotopes of interest and their respective dwell times are listed in Table 2.

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Table 2. Isotopes, dwell times and channels per peak used in this work.

| m/z | Dwell time/ms | Channels/peak | Analyte | Comments |
|-----|---------------|---------------|-------------------------|----------------------------------|
| 228 | 10 | 1 | | Background |
| 235 | 20 | 3 | 235U | Dead time/hydrides/tailing |
| 237 | 30 | 1 | | Tailing |
| 238 | 10 | 3 | ²³⁵ U | Dead time |
| 239 | 30 | 3 | ²³⁸ UH* | Hydrides |
| 245 | 40 | 3 | ²⁴⁵ Cm | Non-irradiated and irradiated Cm |
| 246 | 10 | 3 | ²⁴⁶ Cm | Non-irradiated and irradiated Cm |
| 247 | 40 | 3 | ²⁴⁷ Cm | Non-irradiated and irradiated Cm |
| 248 | 10 | 3 | ²⁴⁸ Cm | Non-irradiated and irradiated Cm |
| 249 | 20 | 3 | ^{2/19} (CI+Bk) | Irradiated Cm |
| 250 | 40 | 3 | ²⁵⁰ Cf | Irradiated Cm |
| 251 | 40 | 3 | ²⁵¹ Cf | Irradiated Cm |
| 252 | 40 | 3 | ²⁵² Cf | Irradiated Cm |

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194 3.2 Peak centre correction

195 The characteristics of Gaussian peaks obtained by mass spectrometers using a quadrupole 196 mass filter [19] limit the accuracy and precision of isotopic ratio measurements. For instance, 197 the measured isotopic ratio could vary gradually over time. For this reason, all isotopic 198 measurements were performed with three channels per peak centred symmetrically around the 199 maximum point (peak centre) (Fig. 3). The peak centre of each isotope was identified visually 200 by performing a scan through the mass range of interest before each measurement. The value 201 of the mass corresponding to the peak centre of each isotope was defined as new mass analyte 202 in the database of the instrument software (PlasmaLab).

As can be seen in figure 3, accurate peak centre correction for ²⁴⁶Cm and ²⁴⁸Cm isotopes improves the accuracy of the non bias corrected ²⁴⁶Cm/²⁴⁸Cm isotopic ratio of the in-house 248 Cm/²⁴⁶Cm = 8.937±0.018 certified solution.



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Figure 3. Peak centre correction for 246Cm and 248Cm isotopes. Three channels per peak were used with a
 separation of ±0.02 a.m.u. from the central channel. (a) Poor symmetrical centring of the channels around the
 peaks centre. (b) Symmetrical centring of the channels around the peaks improves the relative difference
 between the measured non-bias corrected 246Cm/248Cm isotopic ratio (r) and the 248Cm/246Cm isotopic ratio
 reference value (R).

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214 3.2. Dead time correction

For highly accurate measurements, detector dead time was appropriately corrected according to the method proposed by Russ [28] using an isotope ratio of an element differing from unity. To minimize possible dependence between detector dead time and element mass, an isotope ratio of an element with a mass number close to that of the analyte element was selected [29].

For this reason, the isotopic ratio ${}^{235}U/{}^{238}U=0.01014 \pm 0.00001$ of uranium (CRM U010) certified solutions with different concentration levels was determined with the automatic detector dead time correction disabled (set to zero *via* the software). For each concentration level, The 'normalized' isotope ratios (isotope ratio corrected for the dead time divided by the true value) were plotted as a function of the values used for the dead time correction leading to curves with a point of intersection (Figure 4). All dead time corrections were carried out according to equation (1)

227

$$C_{corr} = \frac{C_{meas}}{1 - \tau C_{meas}} \tag{1}$$

229

Where, C_{corr} is the corrected intensity (cps), C_{meas} is the measured intensity and τ is the detector dead time, typically τ =10-50 ns [30, 31]. The intersection of the curves corresponding to different concentration levels provides the correct value for the detector dead time.

In addition the y-position of the intersection point permits an estimation of the amount of mass discrimination. In Figure 4a, it can be seen that the intersection of the curves gives a detector dead-time equal to 34 ns. The same result can be better visualized by plotting the standard deviation of the dead time corrected isotope ratios for the different concentration levels as a function of the applied dead time correction. The dead time is obtained for the minimum value of the standard deviation (Fig. 4b).





Figure 4. Normalized U-235/U-238 isotope ratio (a) and its standard deviation (b) as a function of the applied
 values for dead time correction, obtained for U010 certified reference solutions with a uranium concentration
 ranging from 1 to 4 ppb.

245 3.3. Mass bias correction

A sample-standard bracketing procedure was adopted to correct for mass bias. Instrumental mass bias is significant in ICP-MS analyses and must therefore be carefully determined and corrected. For ICP-MS, heavier isotopes have a better transmission efficiency through the plasma-vacuum interface region, and so isotopic ratios are biased to the heavy isotopes [32]. For mass bias correction, the exponential law was used [33, 34]:

251

$$R = r \left(\frac{m_2}{m_1}\right)^{\beta} \tag{2}$$

253

252

- where r and R are the measured and true ratios of isotope2/isotope1; m_2 , m_1 are atomic 254 255 masses of isotope2 and isotope1, and β is the mass fractionation factor.
- 256 Measurements of both non-irradiated and irradiated Cm samples were bracketed with the in-
- house 248 Cm/ 246 Cm = 8.937±0.018 standard solution. Mass bias corrections were performed in 257
- all isotope ratios assuming identical Cm, Bk, and Cf mass bias factors. 258
- 259
- 260 3.4. Corrections for abundance sensitivity and hydrides formation
- The high intensity beam of ^{246, 248}Cm isotopes during measurement of the concentrated sample 261
- solutions (~100 ppb of ²⁴⁸Cm) creates spectral interferences on the ^{245, 247}Cm and ²⁴⁹(Bk+Cf) 262
- isotopes including hydrides formation and peak tailing. 263
- Peak tailing and hydrides formation for Cm were derived experimentally by using U010 264 265 uranium standard solution assuming similar U and Cm peak tailing and hydrides formation. 266 Measurements of U peak tailing and hydrides formation were performed on different U010 267 concentrations in order to assure a constant peak tailing and hydrides formation in different 268 ion beam intensities of the major isotope.
- The value of 237/²³⁸U ratio was used to derive tail contribution of ²⁴⁸Cm on ²⁴⁷Cm and of 269 ²⁴⁶Cm on ²⁴⁵Cm to correct the measurement ratios of ²⁴⁵Cm/²⁴⁶Cm and ²⁴⁷Cm/²⁴⁶Cm, 270 271 respectively. Vacuum conditions in the analyser influence the degree of tailing effect [34]. For the analyser vacuum value of our ICP-QMS ($\sim 10^{-7}$ mbar), the average abundance sensitivity 272 during all the analytical session of 238 U at m/z=237 was about 1.97±0.02 ppm (Fig. 5).
- 273
- In order to quantify hydrides contribution of 238 U on m/z=239 the 238 UH⁺/ 238 U⁺ ratio was 274 measured. This ratio was used to correct ²⁴⁷Cm and ²⁴⁹(Bk+Cf) from isobaric interferences of 275 ²⁴⁶CmH⁺ and ²⁴⁸CmH⁺ respectively; ²⁴⁷Cm combine the effect of tailing and hydride 276 formation. The most important factors affecting hydride generation are the mode and rate of 277 sample introduction into the plasma [35-38]. The average $^{238}UH^{+/238}U^{+}$ ratio for the 278 279 introduction system that was used in this work (section 2.1) was found to be 35.7±0.1 ppm 280 (Fig. 5).
- 281 Note that for more precise and accurate results for peak tailing and hydride formation only $237/^{235}$ U and 238 UH⁺/ 235 U⁺ ratios were measured and converted to $237/^{238}$ U and 238 UH⁺/ 238 U⁺ 282 using the isotopic ratio ${}^{235}U/{}^{238}U=0.01014 \pm 0.00001$. 283





Figure 5. Abundance sensitivity and hydrides formation as a function of U-238 ion intensity. Error bars are
 expressed as 95% confidence level (2σ) and are within the symbols.

288 3.5. Instrumental background and memory blank

Instrumental background was monitored during samples measurement at m/z=228 (Table 2) and a typical level was <5 cps.

291 For memory blank, a washing procedure was applied after every sample or standard, 292 including a short pre-wash of 2 min 2% HNO₃ + 2% HCl and a 2% sub-boiling HNO₃ wash for 10 min to reduce isotopes intensities. After washing, the intensities of all the relevant 293 294 isotopes were measured. These values were then used as the blank solution background 295 correction for subsequent sample or standard measurements. Typical values of such solution blanks were ²³⁵U<20 cps, 237<10 cps, ²³⁸U<200 cps, ²⁴⁵Cm<10 cps, ²⁴⁶Cm<15 cps, ²⁴⁷Cm<10 296 cps, ²⁴⁸Cm<50 cps, ²⁴⁹(Bk+Cf)<10 cps, ²⁵⁰Cf<10 cps, ²⁵¹Cf<20 cps, ²⁵²Cf<10 cps. The wash 297 298 procedure effectively reduces the intensities to less than 0.01% of the original ion beams.

299

300 3.6 Accuracy and reproducibility of ²⁴⁸Cm/²⁴⁶Cm isotope ratio measurements

The reproducibility and accuracy of 248 Cm/ 246 Cm isotopic ratio in our certified in-house Cm solution was evaluated using a 4 ppb solution. The results, corrected for dead time and mass bias, for two different days of measurements are presented in Figure 6. The relative standard error or internal precision (RSE = RSD/ \sqrt{n} , where *n* is the number of cycles per analysis, n=25), with 95% of confidence level was between 0.2 and 0.3 %. The reproducibility or external precision, expressed as the relative standard deviation (RSD), obtained for measurements on two different days and for 12 measurements was ~ 0.1 %. The accuracy of the 12 measurements mean value compared to the certified value of 8.937±0.018 was within ~
0.9‰ as calculated using equation (3)

310

311

$$Accuracy = \frac{r-R}{R} \times 1000 \tag{3}$$

where *r* and *R* are the measured and reference value of the 248 Cm/ 246 Cm isotopic ratio, respectively.



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Figure 6. Reproducibility of 248Cm⁺/246Cm⁺ standard solution ratio at two different days of measurements
 using ICP-QMS. Dashed and plain lines represent the average value of the 12 measurements and the
 248Cm⁺/246Cm⁺ ratio reference value (8.937), respectively. Shaded area corresponds to the TIMS reference
 value error (±0.018) expressed at 95% confidence level (2σ).

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320 3.7 Isotopic composition for non-irradiated and irradiated Cm samples

The developed analytical procedure described above was applied for measuring isotopic composition of non-irradiated and irradiated Cm samples. All values obtained were corrected for dead time, mass bias, background, and memory blank. Peak tailing and hydrides corrections were performed for the appropriate isotopes as described in section 3.4, only for the concentrated solutions (~100 ng/g of 248 Cm).

326 Measurements of Cm isotopic ratios before and after sample irradiation are presented in figure 7. In order to validate the efficiency of the peak centre, mass bias, and dead time 327 corrections, the ²⁴⁸Cm/²⁴⁶Cm isotopic ratio of non-irradiated and irradiated Cm samples was 328 329 also determined by TIMS using the total evaporation method. As can be seen in figure 7c, a 330 good agreement between ICP-QMS and TIMS methods was obtained. The relative difference 331 of the average values between the two methods is 0.03% and 0.2% for non-irradiated and 332 irradiated Cm samples, respectively. Isotopic composition measurements of Cf and Bk created after Cm irradiation are presented in figure 8. In the present work, isobaric 333 interference at m/z=249 of ²⁴⁹Cf and ²⁴⁹Bk was not resolved. 334





336 Figure 7. Measurements of Cm isotopic composition for non-irradiated (left) and irradiated (right) by ICP-QMS

337 (squares, circles) and TMS (diamonds). Dashed lines represent the average value of the measurements. All errors

338





339

Figure 8. Measurements of Cf and Cf+Bk, isotopic composition. Dashed lines represent the average value of the
 measurements. All errors are expressed at the 95% confidence level (2σ).

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343 The 249 (Cf+Bk)/ 251 Cf isotopic ratio (Fig. 8a) was measured assuming identical Cf and Bk 344 sensitivities. The assumption is realistic considering their first ionization potential [39]. Cf

and Bk separation by chromatographic techniques or eventually with the collision reaction cell of the ICPMS could be attractive methods to resolve this interference over the time scale of the present work. Another method plans for this analysis is the use of liquid scintillation counting for measuring the β -deacy of ²⁴⁹Bk. The average values of isotopic composition for both non-irradiated and irradiated Cm samples are summarized in Table 3.

Uncertainties of all variables including ²⁴⁸Cm/²⁴⁶Cm isotopic ratio, the certified solution, internal precision of measured isotopic ratios, peak tailing, and hydrides formation, were propagated using the general equation for error propagation [40] to determine the total error on the isotope ratios. All errors quoted in this study are at the 95% confidence level (2σ). Isotopic ratio errors ranged from 0.3% to 1.3% (Table 3). Important factors that affect isotopic ratio error are: background corrections for peak tailing and hydrides formation, and low isotopic abundance (intensity beam<2000 cps).

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Table 3. Average isotopic composition values of non-irradiated and irradiated Cm samples.

| Concentration of ²⁴⁸ Cm in ppb | Isotope ratio | Method | Number of replicates | Non-irradiated | % Error | Irradiated | % Error |
|---|---|---------|----------------------|----------------|---------|---------------|---------|
| ~100 | 245Cm+/246Cm+ | ICP-QMS | 3 | 0.010821 (85) | 0.78 | 0.001918 (15) | 0.92 |
| ~100 | 247 Cm ⁺ /246 Cm ⁺ | ICP-QMS | 3 | 0.004687(61) | 1.29 | 0.005389(61) | 1.13 |
| ~4 | 248 Cm ⁺ /246 Cm ⁺ | ICP-QMS | 5 | 38.55(13) | 0.33 | 38.30(11) | 0.29 |
| | | TIMS | 3 | 38.54(11) | 0.28 | 38.23 (9) | 0.24 |
| ~100 | 249(Bk+ + Cf+)/251Cf+ | ICP-QMS | 3 | | | 16.55 (11) | 0.68 |
| ~100 | ²⁵⁰ Cf ⁺ / ²⁵¹ Cf ⁺ | ICP-QMS | 3 | | | 6.387 (34) | 0.54 |
| ~ 100 | ²⁵² Cf ⁺ / ²⁵¹ Cf ⁺ | ICP-QMS | 3 | | | 0.3778 (33) | 0.87 |
| | | | | | | | |

359 360

361 **4.** Conclusion

362 In this work, an analytical procedure for the isotopic measurement of transuranium isotopes 363 (Cm, Cf) based on quadrupole ICP-MS was developed. The accurate and precise isotopic 364 composition determination of Cm sample before and after irradiation was achieved by 365 controlling and correcting factors that affect isotopic accuracy and precision, such as peak 366 centre correction, detector dead time, mass bias, peak tailing, hydrides formation, 367 instrumental background, and memory blank. Isotopic ratio uncertainties of all variables 368 ranged from 0.3% to 1.3%. The precision attained on these measurements with ICP-QMS is 369 sufficient to deduce the actinide capture cross sections with the precision required for nuclear 370 data libraries.

371 Acknowledgments

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The Cm sample was prepared at Lawrence Berkeley National Laboratory (LBNL) under the support of the Office of Sciences, Office of Basic Energy Sciences and the Division of Chemical Sciences, Geosciences and Biosciences of the U.S. Department of Energy under

- 376 Contract No. DE-AC02-05CH11231 at LBNL. The authors are indebted for the use of the
- 377 curium material to the Office of Basic Energy Sciences through the transplutonium element
- 378 production facilities at the Oak Ridge National Laboratory.
- 379

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