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3	ELSEVIER	Appl	lied Radiation and Isotopes	5 I (IIII) III-III	Isotopes	
5	Technical note					
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9	A combined ${}^{148}$ Gd/ ${}^{244}$ Cm source for energy calibration of $\alpha$ -spectrometers					
11		A.F. Novg	gorodov <sup>a</sup> , N.A. k	Korolev <sup>a</sup> , F. Roesch <sup>t</sup>	),*	
13	<sup>a</sup> Joint Institute for Nuclear Research, RUS-141980 Dubna, Russian Federation <sup>b</sup> Institute of Nuclear Chemistry, Johannes Gutenberg-University of Mainz, Fritz-Strassmann-Weg 2, 55128 Mainz, Germany Received 7 April 2005; received in revised form 1 June 2005; accepted 23 August 2005					
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17						
19	Abstract					

#### Abstract

For high-resolution  $\alpha$ -particle spectroscopy, detector calibration sources of highest performance are required concerning thickness and homogeneity. In addition, the  $\alpha$ -emitting radionuclides must have adequate half-lives and  $\alpha$ -emission energies. This paper describes a new type of  $\alpha$ -emitting calibration source, represented by a mixture of <sup>148</sup>Gd and <sup>244</sup>Cm. This system covers an  $\alpha$ -energy range from 3.18 MeV (<sup>148</sup>Gd) to 5.76 and 5.80 MeV (<sup>244</sup>Cm). The source has been prepared by electrolytic deposition and has good performance in terms of

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thickness and homogeneity.
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## 29 **1. Introduction**

For high-resolution α-particle spectroscopy, detector calibration sources of highest performance are required
 concerning thickness and homogeneity. Recent developments focus on chemical aspects of optimal preparation of
 layers for conventional α-emitters (Tsoupko-Sitnikov et al., 2000; Sanchez et al., 2002; Leprince and de Sanoit, 2002).

 $_{37}$  However, there might be further improvements of calibration sources as regards the selection of unique  $\alpha$ -emitter

39 with adequate half-life and, in particular,  $\alpha$ -particle energy. Here we describe a new type of  $\alpha$ -emitting calibration 41 source, consisting of a mixture of <sup>148</sup>Gd and <sup>244</sup>Cm. This

system presents only two strong  $\alpha$ -energies and covers a very broad  $\alpha$ -energy range from 3.18 MeV (<sup>148</sup>Gd) to 5.80 MeV (<sup>244</sup>Cm).

45 State-of-the-art calibration sources for α-spectroscopy such as <sup>226</sup>Ra, <sup>236,238–240</sup>Pu, etc. have several suboptimum
47 properties. Among others, contamination of the detectors might occur due to recoil nuclei or fragmentation of the source material (aggregate recoil). Coating of the source surfaces with thin layers of specific materials might

51 minimize these effects; however, the covering layer might

E-mail address: frank.roesch@uni-mainz.de (F. Roesch).

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decrease the energy resolution (up to 500–100 keV of the 59 FWHM). Moreover, the  $\alpha$ -energy range represented by these sources is rather narrow [<sup>226</sup>Ra:  $T_{1/2} = 1600$  a 61 ( $E_{\alpha} = 4601$  and 4784 MeV); <sup>236</sup>Pu:  $T_{1/2} = 2.85$  a ( $E_{\alpha} = 5721$  and 5768 MeV); <sup>238</sup>Pu:  $T_{1/2} = 87.74$  a ( $E_{\alpha} = 5457$  and 63 5499 MeV); <sup>239</sup>Pu:  $T_{1/2} = 2.41 \times 10^4$  a ( $E_{\alpha} = 5143$  and 5156 MeV); <sup>240</sup>Pu:  $T_{1/2} = 6563$  a ( $E_{\alpha} = 5124$  and 65 5168 MeV)].

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## 2. Materials and methods

71 The source proposed consists of a combination of two individual,  $\alpha$ -emitting radionuclides in one uniform source. 73 These radionuclides are <sup>148</sup>Gd and <sup>244</sup>Cm. Their decay data are summarized in Table 1. The proposed mixture provides 75 a very reliable energy calibration, because it covers an  $\alpha$ energy range of almost 4 MeV. The <sup>148</sup>Gd decays to a 77 stable, non-radioactive daughter ( $^{144}$ Sm), while the decay of  $^{244}$ Cm results in the formation of  $^{240}$ Pu, which has a 79 rather long physical half-life of 6563 a. Thus, the radioactivity of <sup>240</sup>Pu created by the decay of <sup>244</sup>Cm is negligible 81 compared to the radioactivity of the calibration source itself. 83

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<sup>53 \*</sup>Corresponding author. Tel.: +49 6131 392 5321; fax: +49 6131 392 4692.

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1	Table	1

Decay characteristics (	Firestone et al	1996) of the $\alpha$ -emi	tters involved
Decay characteristics	Thestone et al.	1990) of the <i>u</i> -chi	tiers involved

Initial radionu	Initial radionuclides			Decay products			
Radionuclide	$T_{1/2}$ (a)	$E_{\alpha}$ (keV)	α-branch (%)	Radionuclide	$T_{1/2}$ (a)	$E_{\alpha}$ (keV)	α-branch (%)
<sup>148</sup> Gd	74.6	3182.680(24)	100	<sup>144</sup> Sm	Stable		
<sup>244</sup> Cm	18.10	5804.77(5)	76.4(12)	<sup>240</sup> Pu	6563	5123.45(23)	26.39(21)
		5762.16(3)	23.6(12)			5168.13(15)	73.51(36)
		Others	< 0.03			Others	< 0.1

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## 2.1. Radionuclide availability

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The <sup>148</sup>Gd was produced in (p, spall) processes of Ta 17 targets at CERN, Geneva, in 2000. Following radiochemical separation of the massive tantalum target at the

19 Institute of Nuclear Chemistry at the University of Mainz, Germany, in 2001, the gadolinium fraction of the neutron-

21 deficient lanthanides was isolated (Novgorodov et al.,

2002). Within 3 years, the initial content of  $^{151}$ Gd ( $T_{1/2}$ ) 23  $_2 = 120$  d) and  $^{153}$ Gd ( $T_{1/2} = 241.6$  d) decayed, and only a pure fraction of  $^{148}$ Gd remained.  $^{244}$ Cm is commercially 25 available.

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## 29 2.2. Source preparation

31 The individual radionuclide solutions were mixed in different ratios (see below) and used for the source 33 preparation. The sources were produced from small

- volumes of 1-3 ml of 0.1 N Na<sub>2</sub>SO<sub>4</sub>/0.01 N H<sub>2</sub>SO<sub>4</sub> by 35 electrolytic deposition on stainless steel (type 1810)
- cathode. The cathode current density was set to 37 80–100 mA/cm<sup>2</sup>. The deposition time was 1-2h. The chemical yield of this process was 95-99%. The quality
- 39 of the prepared sources is demonstrated in Fig. 1, indicating the excellent energy resolution of the main  $\alpha$ -
- 41 groups. The FWHM are 14.8 keV for <sup>148</sup>Gd and 15.4 keV for <sup>244</sup>Cm.
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## 2.3. Mixtures

According to the different branches of the individual  $\alpha$ -49 emissions (cf. Table 1), the two radionuclides were mixed initially in an absolute activity ratio of <sup>148</sup>Gd:<sup>244</sup>Cm of

51 1:1.3. This resulted in comparable peak areas of the  $E_{\alpha} = 3183 \text{ keV}$  line of <sup>148</sup>Gd and the 5805 keV line of 53 <sup>244</sup>Cm. The energy spectrum of the sources is presented in

Fig. 1, representing the  $\alpha$ -spectrum of a source of 300 Bq 55  $^{148}$ Gd and 400 Bq  $^{244}$ Cm. The area of the detector used was

 $1 \text{ cm}^2$ . The distance between source and detector was 3 cm.

57 The measurement time was 965 s.

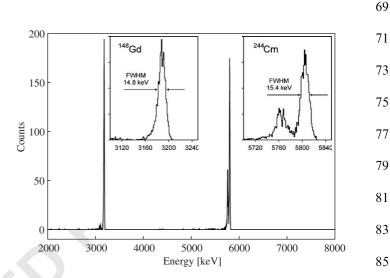


Fig. 1.  $\alpha$ -spectrum of a source of 300 Bq <sup>148</sup>Gd and 400 Bq <sup>244</sup>Cm representing the whole range of  $\alpha$ -energies between 2 and 8 MeV. The area of the detector used was 1 cm<sup>2</sup>. The distance between source and detector was 3 cm. The measurement time was 965 s. The insets show the strongest lines on a magnified energy scale.

#### 3. Discussion

The measured  $\alpha$ -spectrum of the combined source of 95 <sup>148</sup>Gd (300 Bq) and <sup>1244</sup>Cm (400 Bq) clearly indicates the high resolution of the main  $\alpha$ -particle groups of FWHM 97 14.8 keV for <sup>148</sup>Gd and 15.4 keV for <sup>244</sup>Cm and the broad range of about 4 MeV, covered by this unique mixture. It 99 thus represents an almost ideal type of  $\alpha$ -calibration source.

Provided the shelf life of this source is assumed to be 101 three half-lives of the shorter-lived radionuclide <sup>244</sup>Cm, this source will be useful for more than 50 years. The recoil 103 nuclei activity arising from <sup>240</sup>Pu hitting the detector surface during a regular calibration procedure (e.g. 900s, 105 4000 Bq of <sup>244</sup>Cm,  $\Theta = 3\%$ ) will correspond to 36 nBq. This will not cause any influence on subsequent determina-107 tions of real samples (e.g. <sup>240</sup>Pu or others from environmental probes), at least for more than 1000 calibration 109 measurements for one detector. Moreover, the aggregate recoil will not affect the determination of plutonium and 111 americium radioisotopes, because their a-energies are inbetween the ones of the calibration source constituents. 113

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 In conclusion, the combined <sup>148</sup>Gd and <sup>244</sup>Cm α-energy calibration sources might be widely used for routine
 detection of α-radiations both in fundamental investiga-

- tions in nuclear physics and nuclear chemistry as well as inapplied research such as environmental analysis.
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