J Radioanal Nucl Chem (2015) 304:1359–1363
DOI 10.1007/s10967-015-4001-0

TANDEM: a mutual cooperation effort for transactinide nuclear data evaluation and measurement

M. Rossbach · C. Genreith · T. Randriamalala · E. Mauerhofer · Zs. Revay · P. Kudejova · S. Söllradl · T. Belgya · L. Szentmiklosi · R. B. Firestone · A. M. Hurst · L. Bernstein · B. Sleaford · J. E. Escher

Received: 23 December 2014 / Published online: 15 February 2015
© Akadémiai Kiadó, Budapest, Hungary 2015

Abstract The need for accurate nuclear reaction data of actinides is well documented and several initiatives from international organizations for improvement have been initiated in the past. This need, particularly in view of method development for non-destructive assay of nuclear waste, has generated a joint effort to use prompt and delayed neutron activation techniques to enhance nuclear capture data of some long lived actinides such as ^{237}Np, ^{242}Pu and ^{241}Am in the frame of a multilateral cooperation. This research initiative is targeted to lay grounds for the development of a non-destructive active neutron interrogation technique to quantify actinides in mixed waste and residues from decommissioning of nuclear installations for safe treatment and storage of such materials.

Keywords Prompt gamma activation analysis (PGAA) · Actinides · Neutron capture cross section · Fission neutrons · Inelastic scattering reactions · Gamma-ray spectroscopy

M. Rossbach (✉) · C. Genreith · T. Randriamalala · E. Mauerhofer
Institute of Energy and Climate Research, IEK-6: Nuclear Waste Management and Reactor Safety, Forschungszentrum Jülich GmbH (FZJ), 52425 Jülich, Germany
e-mail: m.rossbach@fz-juelich.de

Zs. Revay · P. Kudejova · S. Söllradl
Technische Universität München, Forschungsneutronenquelle Heinz Maier-Leibnitz, FRM II, 85748 Garching, Germany

T. Belgya · L. Szentmiklosi
Centre for Energy Research, Hungarian Academy of Sciences, (MTA EK), Budapest, Hungary

R. B. Firestone · A. M. Hurst
Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

L. Bernstein
Department of Nuclear Engineering, University of California, Berkeley, CA 94720-1500, USA

B. Sleaford · J. E. Escher
Lawrence Livermore National Laboratory, Livermore, CA 94551, USA

Introduction

The International Atomic Energy Agency (IAEA) and the Nuclear Energy Agency (NEA) of the OECD have pointed out at various opportunities that actinide nuclear data are scarce and sometimes rather uncertain [1–4]. Initiatives by the EU have been launched to enhance international cooperation for improvement of actinide nuclear data such as TALISMAN (Transnational Access to Large Infrastructure for a Safe Management of Actinides) [5], ERINDA (European Research Infrastructures for Nuclear Data Applications) or CHANDA (Solving Challenges in Nuclear Data for the Safety of European Nuclear Facilities) [6]. The benefit of these programs rests largely in providing access to necessary infrastructures, however, cooperative research in the field of method development and actinide characterization is strongly dependent on individual initiatives. Hence, the three groups working on prompt gamma activation analysis (PGAA) at the FZJ, the FRM II and the MTA EK agreed to sign a Memorandum of Understanding (MoU) in 2011 and initiated the TANDEM collaboration. The intention of both is to explore and develop PGAA as a tool for improving nuclear reaction data of actinides with a focus on nuclear waste and related issues. At a later stage the LBNL and LLNL joined the group as research interests coincided and actinides of interest could be made available

for joint experiments at the neutron beam facilities in Budapest and Garching. 3 years of successful cooperation have generated various results and also plans to enhance the focus of TANDEM into the investigation of long lived fission products are being pursued.

Materials and methods

PGAA using thermal or cold neutron beams at high-flux research reactors like Forschungsneutronenquelle Heinz Maier-Leibnitz, FRM II in Garching, Germany, is an ideal tool for basic nuclear prompt and decay investigations as it provides well-defined neutron energy, sample irradiation conditions in a low background environment, together with sensitive and versatile detector and counting conditions. The major constraint experienced at FRM II is limited availability of beam time as worldwide there are only a few PGAA facilities offering high enough neutron fluxes. For our experiments we could also use the Budapest reactor PGAA system offering a lower thermal equivalent neutron flux intensity of 7×10^7 n cm^{-2} s^{-1} compared to FRM II with 2×10^{10} n cm^{-2} s^{-1}. Both experimental facilities are described in more detail in [7–9].

Sample preparation techniques for actinides have been developed at the Institute IEK-6 of the Forschungszentrum Jülich GmbH [10, 11]. 4 × 4 cm Suprasil© quartz sheets (0.2 ± 0.02 mm thick) have been purchased from Heraeus Quarzschmelze Hanau, Germany. ^{237}Np (isotopically pure) and ^{242}Pu (99.93 % enriched) were obtained from Oak Ridge National Laboratory (ORNL) via Technische Universität München (TUM) as oxides and pressed into pellets of 3 mm diam. (2.3–8.2 mg) The samples were encased between the quartz sheets and sealed with epoxy. ^{241}Am activity (185 MBq) was obtained from Eckert & Ziegler, Nuclitec, Braunschweig and delivered to PTB in Braunschweig, the German National Metrology Institute, for accurate preparation and calibration of the sources. A drop of the ^{241}Am solution was dried on the centre of one of the quartz sheets and afterwards covered with another sheet. Two samples were prepared by drying the droplet on top of small circular gold foils (3 mm diam., 0.003 mm thick, 0.4 mg), serving as neutron flux monitors. These were also sealed between quartz sheets using epoxy. The activity of the ^{241}Am in the samples was determined with an uncertainty of 0.75 %, hence the mass of ^{241}Am could be calculated with similar accuracy.

The photo-peaks in the obtained gamma-ray spectra were evaluated using Hypermet-PC [12–14]. This program was especially developed for the evaluation of complex prompt-gamma spectra in the energy range from 20 keV to 12 MeV. As the resolution of HPGe detectors degrades rapidly at higher energies it is imperative to use software with accurate energy and efficiency calibration as well as correct peak fitting over the entire range of energies and peak shapes encountered. The influence of neutron and gamma attenuation in the samples and casing material was accounted for by careful MCNP and GEANT 4 simulations. Both irradiation and counting facilities for PGAA measurements were simulated for optimization and background reduction prior to the measurements.

DICEBOX [15], a Monte Carlo computer code originally developed at Řež, Czech Republic, generates simulated neutron-capture decay schemes based on nuclear level density and photon-strength function models. The simulated intensities of transitions populating low-lying levels can be normalized to the experimental cross sections de-exciting those levels in order to determine the unobserved cross section feeding the ground state. Combined with the observed cross section feeding the ground state this gives the total radiative thermal neutron-capture cross section σ_0 [16]. This program was applied at LBNL to simulate unobserved transitions in level schemes for ^{242}Pu successfully and will also be used to investigate ^{241}Am and ^{237}Np data.

In the frame of the TANDEM collaboration and with financial support of the Bundesministerium für Bildung und Forschung (BMBF) a new instrument using fission neutrons for PGAA investigations has been developed and applied to investigate inelastic scattering reactions of actinides and other elements. This instrument is composed of a well-shielded HPGe detector with associated electronics at the fast neutron beam at the SR10 beam line of the FRM II reactor. In contrast to cold neutron PGAA monitoring neutron capture (n,γ) reactions, this device will provide information on inelastic scattering (n,n'γ) and perhaps on induced fast fission reactions of the originally exposed isotopes. Decay measurements of the irradiated samples can provide information on other reaction channels, such as (n,p), (n,α) or (n,2n) resulting from the higher energy neutron irradiation. However, for the development of a possible non-destructive analytical method using neutron generators this (n,n'γ) reaction is of particular interest.

Results

Appropriate sample preparation techniques for PGAA investigations of radioactive samples were developed at the Forschungszentrum Jülich. The samples have been irradiated at the PGAA stations in Budapest and Garching and the spectra were evaluated at the FZJ. Thermal capture cross sections were calculated after neutron flux determination from a co-irradiated Au foil (0.003 mm thick). Results for ^{237}Np, ^{242}Pu and ^{241}Am compare very well with literature values, all of which (exception: Belgya et al.

2012) were measured using neutron activation analysis as can be seen from Figs. 1, 2 and 3 [17–20].

As the uncertainties of the experimentally determined cross section for ^{242}Pu are dominated by the relatively large uncertainty of tabulated emission probabilities (not the counting statistics), this nuclide was investigated using DICEBOX at the LBNL, Berkeley. One of the co-workers from Jülich (C. G.) had the opportunity to spend a 2-month scientific visit there to be introduced in using this Monte-Carlo code. Decay transitions from the continuum could be evaluated and added to the experimentally-determined prompt gamma lines. This evaluation is independent from tabulated emission probabilities which are associated with large uncertainties propagating to the experimentally determined values (see Fig. 4). A total radiative thermal neutron capture cross section of 21.9 ± 1.5 b is obtained for the ^{242}Pu (n,γ) reaction from the sum of measured gamma-ray yields directly feeding the ground state in the prompt-gamma analysis, combined with the simulated contribution from the unresolved quasi continuum. Similar evaluations for ^{237}Np and ^{241}Am are in progress.

^{241}Am thermal capture cross section measurements are particularly difficult with NAA techniques as a low energy resonance can affect the 0.0253 eV cross section. Using thermal neutrons a Westcott correction factor needs to be applied to correct for epithermal contributions. As we were using cold neutrons of <10 meV and referencing to the Au 0.0253 eV cross section our values tend to represent the

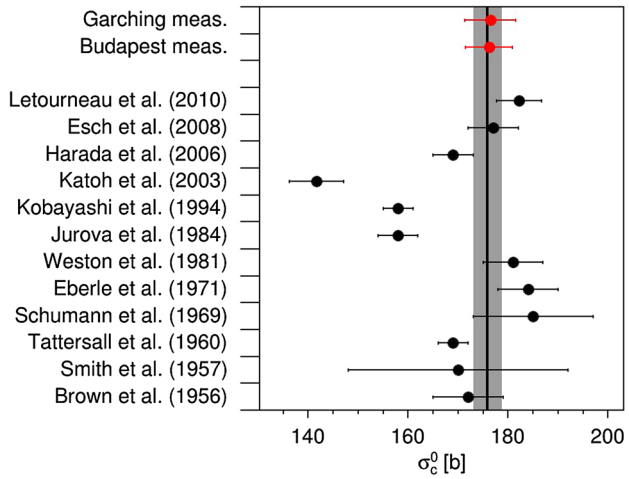

Fig. 1 Neutron capture cross section values for ^{237}Np compared to literature values. The *vertical line* represents the ENDF value

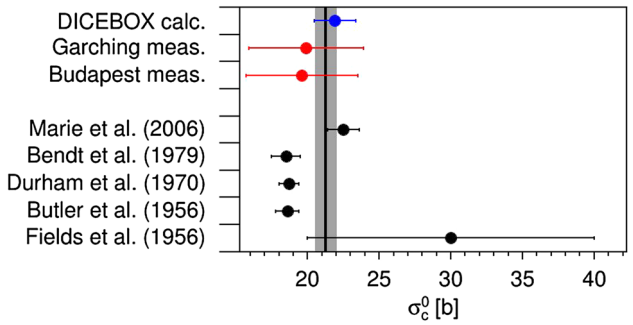

Fig. 2 Neutron capture cross section values for ^{242}Pu compared to literature values. The *vertical line* represents the ENDF value

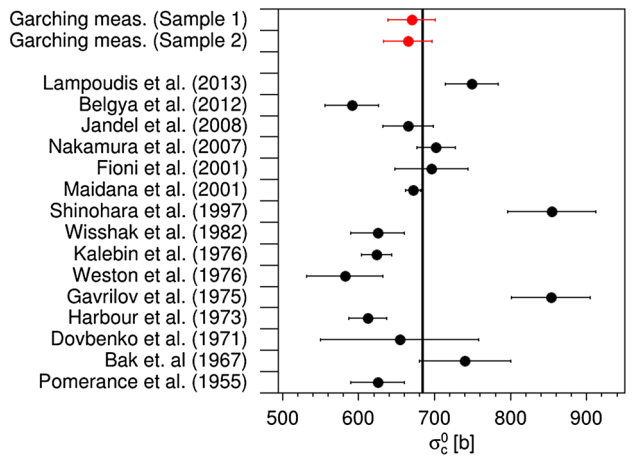

Fig. 3 Neutron capture cross sections values for ^{241}Am compared to literature values. The *vertical line* represents the ENDF value

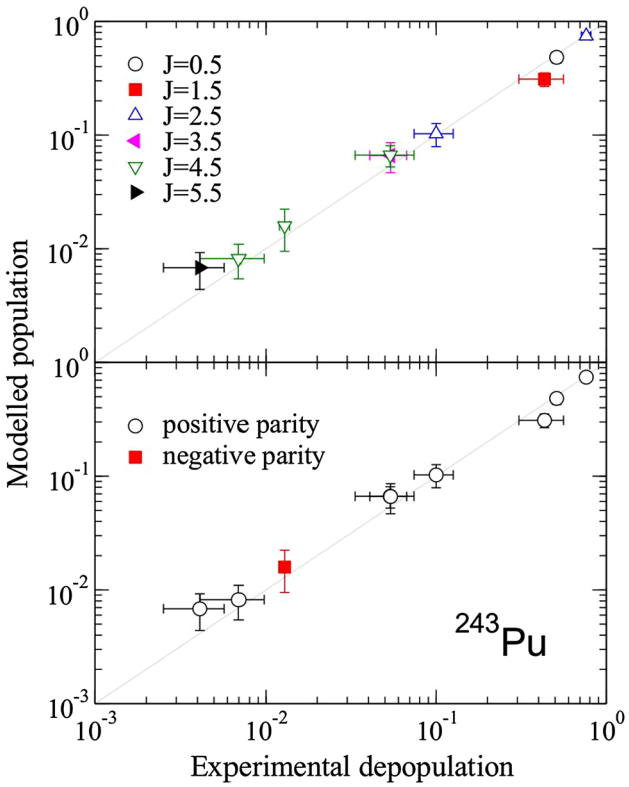

Fig. 4 Comparison between experimental and simulated results for ^{243}Pu using DICEBOX

Fig. 5 FaNGaS, the fast neutron gamma spectroscopy instrument at the SR10 beam line of FRM II Research Reactor in Garching

true 1/v cross section of the ^{241}Am (n,γ) ^{242m,g}Am reaction [19].

With respect to development of PGAA based analysis of actinides the absolute detection limits for ^{237}Np, ^{241}Am and ^{242}Pu under FRM II irradiation conditions are (using low-energy gamma lines up to 300 keV) 0.06, 0.02 and 0.2 μg respectively, and 1.4, 0.6 and 10 μg for high energy gamma rays. These values seem to suffice for the analysis of small samples, such as safeguards swipe samples, and look rather promising for the further development of methods for actinide analysis in e.g. residues from dismantling activities or in low and medium activity nuclear waste forms. Development of innovative fuel and storage matrices for actinides can also profit from a sensitive neutron interrogation technique.

The development and installation of a Fast Neutron Prompt Gamma-Ray instrument at the SR10 beam line of the FRM II is a major achievement of the successful collaboration within the TANDEM cooperation (see Fig. 5). Up to 2.3 × 10^8 cm^{-2} s^{-1} fission neutrons are extracted from a 95 % enriched ^{235}U converter and directed through the S10 beam tube into the bunker shared by the MEDAPP (mainly used for medical applications) and NECTAR (-mainly used for fission neutron imaging) instruments. A specially designed pair of collimators reduces the beam size to 5 cm diameter, which is crucial to decrease the detector background. An electrically cooled 50 % eff. *n*-type HPGe detector (Ortec) is heavily shielded against fast and thermal neutrons as well as against gamma radiation by 15 cm lead, 1 cm B_4C and 30 cm of PE (from the detector to the outside). The HPGe detector is connected to a DSPEC digital spectrum analyser to record the gamma-ray spectra. In front of the 5 cm diameter collimator the samples are mounted in an aluminium frame (50 cm from the detector end cap) for exposure to the neutron beam. Subsequently, the irradiated samples can be measured for decay gammas of produced radioisotopes at a "close-to-detector" position, 15 cm from the detector end cap. Scattered fast neutrons from the sample are shielded by a 6 cm PE plug inside the collimator and a thin ^{6}Li-containing glass in front of the HPGe-detector. This was necessary to reduce neutron damage of the detector and reduced greatly the Ge triangles in the spectra. Careful neutron-flux characterization using the threshold reaction technique is in progress. After checking the feasibility under the actual background conditions, a set of stable elements were irradiated and obtained results compared with the only comprehensive compilation available of inelastic scattering reactions of 1978 [20]. Once the new instrument has demonstrated its suitability and sensitivity our previously prepared actinide samples (^{237}Np, ^{242}Pu, ^{241}Am) will be investigated in detail to extract further information for the development of analytical tools for characterization of nuclear materials. First results obtained with FaNGaS will be described in a forthcoming publication.

Discussion and conclusion

Bringing together a competent team of experts under a common heading, formalized at least to some extent, releases substantial synergies provided a spirit of team work and collaboration prevails. In our case there was substantial exchange of basic knowledge as the individual partners came from different backgrounds; some were more familiar with generation of nuclear data, some were experts in applying the PGAA technique and others could provide experience in sample preparation and/or MCNP simulations. This combination of a suite of capabilities rendered the TANDEM cooperation highly productive and innovative. Data generated by this collaboration will be feed into the evaluation system of LBNL and an attempt will be made to evaluate and improve the Demidov data for inelastic scattering reactions from 1976 [21]. Results for partial capture and thermal neutron capture cross sections will be published in separate reports.

The MoU on PGAA development will be renewed for another 3 years and new partners will be incorporated in the TANDEM cooperation. The focus of our collaboration will be extended to include also long lived fission products such as ^{93}Zr, ^{135}Cs, ^{107}Pd or ^{129}I to the scope of our investigations. Furthermore, it is also a major intent of the collaboration to contribute to the preservation of nuclear basic knowledge by, e.g., training PhD students and introducing postdocs into the fields of PGAA and the physics of activation analysis. The long-range target, however, remains the development of analytical techniques based on neutron active interrogation and gamma spectrometry for

the non-destructive (possibly also remote) characterization of nuclear waste and residues from dismantling activities. These activities will strongly increase in the near future (especially in Germany) and our initiative is particularly motivated to address the need for safe disposal of hazardous materials.

Acknowledgments Many individuals have actively contributed to the success of this collaboration. We would like to sincerely thank them all without mentioning all of them. Special thanks are going to the workshop team of Ayhan Egmen of the FZJ and the team of the FRM II, radiation safety and security. We are particularly thankful for generous financial support by the BMBF under Grant 02S9052 and to Dr. M. Weigl from the Projektträger Karlsruhe as the competent and always helpful administrator of the project. Financial support for our irradiations at the Budapest research reactor was generously provided by the EURATOM FP7 ERINDA project (Grant Agreement No. 269499). This work was performed in part under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344.

References

1. Smith DL, Otuka N (2012) Experimental nuclear reaction data uncertainties: basic concepts and documentation. Nucl Data Sheets 113:3006–3053
2. Forrest RA (2011) Nuclear science and data needs for advanced nuclear systems. Energy Procedia 7:540–552
3. INDC International Nuclear Data Committee: Minor Actinide Reaction Data (MANREAD), International Atomic Energy Agency, Vienna Austria (2010), INDC(NDS)-585
4. Harada H, Plompen A (2014) Meeting nuclear data needs for advanced reactor systems, Report by the Working Party on International Data Evaluation Co-operation of the NEA Nuclear Science Committee, OECD 2014, NEA/NSC/WPEC/DOC(2014)446
5. Project description: www.talisman-project.eu. Accessed Dec 2014
6. Project description: www.chanda-nd.eu. Accessed Dec 2014
7. Kudejova P, Meierhofer G, Zeitelhack K, Jolie J, Schulze R, Türler A, Materna T (2008) J Radioanal Nucl Chem 278:691–695
8. Canella L, Kudejova P, Schulze R, Türler A, Jolie J (2011) Characterisation and optimisation of a new prompt gamma-ray activation analysis facility (PGAA) at FRM II. Nucl Instrum Methods A 636:108–113
9. Szentmiklosi L, Belgya T, Revay Zs, Kis Z (2010) J Radioanal Nucl Chem 286:501–505
10. Genreith C, Rossbach M, Mauerhofer E, Belgya T, Caspary G (2012) Nukleonika 57:443–446
11. Genreith C, Rossbach M, Mauerhofer E, Belgya T, Caspary G (2013) J Radioanal Nucl Chem 296:699–703. doi:10.1007/s10967-012-2080-8
12. Phillips GW, Marlow KW (1976) Program HYPERMET for automatic analysis of gamma-ray spectra from Ge detectors. Report of NRL progress, Issue May, pp 40–41
13. Fazekas B, Belgya T, Dabolczi L (1996) HYPERMET-PC: program for automatic analysis of complex gamma-ray spectra. J Trace Microprobe Tech 14:167–172
14. Révay Z, Belgya T, Molnár G (2005) J Radioanal Nucl Chem 265:261–265
15. Bĕcvář F (1998) Nucl Instrum Methods A 417:434
16. Firestone RB, Abusaleem K, Basunia MS, Bečvář F, Belgya T, Bernstein LA, Choi HD, Escher JE, Hurst AM, Krtička MM, Revay Z, Rogers AM, Rossbach M, Siem S, Sleaford B, Summers NC, Szentmiklosi L, van Bibber L, Wiedeking M (2014) EGAF: measurement and analysis of gamma-ray cross sections. Nucl Data Sheets 119:79–87 (In Proceedings of International Conference on Nuclear Data Science and Technology, March 4–8, 2013 New York, USA)
17. ENDF/B-VII.1 (2011) https://www-nds.iaea.org/exfor/endf.htm. Accessed Nov 2013
18. Genreith C, Rossbach M, Révay Z, Kudejova P (2013) Determination of (n, γ) cross sections of 241Am by PGAA. Nucl Data Sheets 119:69–71 (In Proceedings of International Conference on Nuclear Data Science and Technology, March 4–8, 2013 New York, USA)
19. Rossbach M, Genreith C (2013) ^{241}Am: a difficult actinide for (n,γ) cross section measurement. In: Proceedings of the ERINDA Workshop, CERN, Geneva, Switzerland, 1–3 October 2013, Chiaveri E (ed), CERN-Proceedings-2014-002, Geneva, pp 157–163
20. Genreith C (2014) Partial neutron capture cross sections of actinides using cold neutron prompt gamma activation analysis. PhD Thesis, Rheinisch Westfälische Technische Hochschule RWTH Aachen
21. Ahmed MR, Al-Najjar S, Al-Amili MA, Al-Assafi N, Rammo N, Demidov AM, Govor LI, Cherepantsev YuK (1978) Atlas of gamma-ray spectra from the inelastic scattering of reactor fast neutrons. Atomizdat, Moscow