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# **INDC International Nuclear Data Committee**

## **Summary Report**

### **Technical Meeting on**

### **Intermediate-term Nuclear Data Needs for Medical Applications: Cross Sections and Decay Data**

IAEA Headquarters  
Vienna, Austria  
22-26 August 2011

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September 2011

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**Abstract**

A summary is given of a Technical Meeting on “Intermediate-term Nuclear Data Needs for Medical Applications: Cross Sections and Decay Data”. Participants assessed and reviewed cross-section and decay data for an extensive number of radionuclides deemed as potentially suitable for application in nuclear medicine and radiotherapy. Technical discussions are described in this report, along with comprehensive listings and detailed recommendations for future work.

Recommendations focused on cross-section studies for a reasonably wide range of targets and projectiles, along with decay data measurements for specific radionuclides. Cross-section and decay-data evaluations are also merited to ensure the necessary quality and consistency of the data to be assembled in an appropriate IAEA-NDS database.

September 2011



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## 1. INTRODUCTION

Diagnostic and therapeutic nuclear medicine extends across a wide range of medical activities in an efficacious manner for the benefit of human health. Many useful or potentially useful radionuclides have been identified for various life-saving applications. The production routes and decay properties of all such radionuclides need to be well defined, but there are remaining deficiencies, especially with regard to the optimum production of specific radionuclides, the minimization/elimination of impurities, and the adequate quantification of various decay parameters (e.g., half-life, and alpha-particle, electron, positron, gamma-ray and X-ray emission probabilities for important dose calculations).

Cyclotrons and accelerators are being used in an increasing number of countries to produce radionuclides for both diagnostic and therapeutic purposes, along with reactors employed to produce specific activation and fission products. Nuclear data are required for both accelerator and reactor production. The current status of such work as organised under the auspices of the IAEA was described in detail by R. Capote. Nuclear data needs were initially addressed by a Coordinated Research Project (CRP) on “Charged Particle Cross-Section Database for Medical Radioisotope Production: Diagnostic Radioisotopes and Monitor Reactions” that concluded in 2001 with the publication of IAEA-TECDOC-1211<sup>1</sup>. Equivalent requirements for the production of therapeutic radionuclides were addressed through a further CRP on “Nuclear Data for the Production of Therapeutic Radionuclides” which started in 2003 and was completed in 2007. The CRP produced a much needed database and handbook<sup>2</sup> covering reactions used for medically important therapeutic radionuclides. The database containing data from both CRPs is available on:

<http://www-nds.iaea.org/medportal/>

Recommended cross-sections were sufficiently accurate to meet the demands of all current applications at the time. The improved quality of the nuclear data that was generated during those CRPs made radionuclide production much more efficient and also enhanced nuclide quality through improved purity.

Following on from the above studies, a consultants’ meeting was held on “High-Precision Beta-Intensity Measurements and Evaluations for Specific PET Radioisotopes” from 3 to 5 September 2008 at IAEA Headquarters, Vienna, Austria. Participants assessed and reviewed the decay data for about 50 positron-emitting radionuclides, and recommended a series of measurements and evaluations to improve the known decay characteristics of these existing or potential PET nuclides<sup>3</sup>.

A further consultants’ meeting was held on “Improvements in Charged-Particle Monitor Reactions and Nuclear Data for Medical Isotope Production” from 21 to 24 June 2011, at the

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<sup>1</sup> **IAEA-TECDOC-1211**, “Charged Particle Cross-Section Database for Medical Radioisotope Production: Diagnostic Radioisotopes and Monitor Reactions”, IAEA Technical Report, Vienna, May 2001. Available online: <http://www-nds.iaea.org/publications/tecdocs/iaea-tecdoc-1211.pdf>

<sup>2</sup> “Nuclear Data for the Production of Therapeutic Radionuclides”, E. Běták, A.D. Caldeira, R. Capote, B.V. Carlson, H.D. Choi, F.B. Guimarães, A.G. Ignatyuk, S.K. Kim, B. Kiraly, S.F. Kovalev, E. Menapace, M. Nortier, P. Pompeia, S.M. Qaim, B. Scholten, Yu.N. Shubin, J.-Ch. Sublet, F. Tárkányi and A.L. Nichols, **IAEA Technical Reports Series No. 473**, 2011, Editors: S.M. Qaim, F. Tárkányi and R. Capote, International Atomic Energy Agency, Vienna, Austria, ISBN 978-92-0-115010-3.

<sup>3</sup> R. Capote and A.L. Nichols, Consultants’ Meeting on “High-precision beta-intensity measurements and evaluations for specific PET radioisotopes”, 3-5 September 2008, IAEA report **INDC(NDS)-0535**, December 2008, IAEA, Vienna, Austria.

same venue<sup>4</sup>. Specific recommendations from both of these consultants' meetings were brought together in June 2011 to formulate and agree the scope, work programme and deliverables of a Coordinated Research Project (CRP) designed to focus on improvements in charged-particle monitor reactions and nuclear data for medical radionuclides (see IAEA report INDC(NDS)-0591, August 2011<sup>4</sup>).

Continued developments in medical imaging and therapy utilizing novel diagnostic and therapeutic techniques and the production of new radionuclides call for a detailed review and further expansion of the database over an intermediate-term timescale defined as between 5 and 10 years (up to approximately 2022/25). All relevant nuclear data need to be critically reviewed, and new measurements and evaluations recommended if necessary. Therefore, a Technical Meeting on "Intermediate-term Nuclear Data Needs for Medical Applications: Cross Sections and Decay Data" was held at IAEA Headquarters, Vienna, Austria, from 22 to 26 August 2011, in order to fulfil these requirements.

M. Venkatesh (Director, NAPC) welcomed the participants. She emphasized the significance of their role in assessing the nuclear data needs for the identification, preparation and characterisation of radionuclides to be used in future medical applications. The primary objective of the meeting should be to assist in defining a series of work programmes to improve preparative procedures, radionuclidic purity, and the quantification of various important decay characteristics. Eight external consultants attended (S.M. Qaim, M.-M. Bé, B.V. Carlson, F.G. Kondev, O. Lebeda, A.L. Nichols, G.F. Steyn and S. Takács) and the IAEA was represented by R. Capote (Scientific Secretary, Nuclear Data Section). S.M. Qaim (Forschungszentrum Jülich, Germany) was elected Chairman of the meeting, and A.L. Nichols (University of Surrey, Guildford, UK) served as rapporteur. The approved Agenda is attached (Appendix 1), as well as a list of participants and their affiliations (Appendix 2).

## 2. PRESENTATIONS

Presentations by the consultants are available on IAEA-NDS web page <http://www-nds.iaea.org/nds-technical-meetings/TM-Med-Apps-2011/>.

### 2.1. Nuclear Data for Medical Applications: An Overview, S.M. Qaim (Forschungszentrum Jülich, Germany)

A brief overview was given of nuclear data research for medical applications, with particular reference to the work carried out at the Research Centre Jülich over the previous three decades. Work has concentrated mainly on the development of accelerator-produced radionuclides, which find application in both diagnosis and internal radiotherapy. Some cross-section measurements have also been performed with respect to external radiation therapy. With respect to radionuclides, both their radioactive decay data and formation cross sections via nuclear reactions are of great significance [1.1, 1.2]. Decay data are needed to calculate the internal radiation dose, which is the deciding factor in the medical use of a radionuclide. Nuclear reaction cross-section data, on the other hand, offer the possibility to optimise a production process (i.e., maximise the yield of the desired product and minimise that of the impurity).

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<sup>4</sup> R. Capote and F.M. Nortier, Consultants' Meeting on "Improvements in charged-particle monitor reactions and nuclear data for medical isotope production", 21-24 June 2011, IAEA report **INDC(NDS)-0591**, August 2011, IAEA, Vienna, Austria. Available online at <http://www-nds.iaea.org/publications/indc/indc-nds-0591.pdf>



Extensive worldwide efforts have been invested to determine with confidence the nuclear data for the most commonly used radionuclides. Detailed cross-section studies have been performed at Jülich on the diagnostic radionuclides  $^{123}\text{I}$  and  $^{201}\text{Tl}$  (for SPECT) and  $^{11}\text{C}$  and  $^{18}\text{F}$  (for PET). Similarly, the therapeutic radionuclides  $^{67}\text{Cu}$ ,  $^{75}\text{Se}$ ,  $^{77}\text{Br}$  and  $^{103}\text{Pd}$  have also been extensively investigated. Both decay data and reaction cross-section data are now fairly well known for the commonly used radionuclides. More specifically under guidance from Jülich, the IAEA has taken the lead in the standardisation of cross-section data. Such data for diagnostic radionuclides were evaluated about 10 years ago (IAEA-TECDOC-1211), and the more recent completion of further IAEA-CRP has created a strong database of therapeutic radionuclides (IAEA Technical Reports Series No. 473, submitted for publication).

Recent nuclear data work has been related to the development of (a) novel positron emitters, and (b) radionuclides for internal therapy, especially those emitting low-energy, highly-ionising radiation. Most of the production-related work on novel positron emitters has been undertaken at Jülich (partly in collaboration with ATOMKI), using highly-enriched targets and low-energy cyclotrons involving mainly the (p,n) reaction. A total of about twenty-five novel positron emitters have been developed, and the database is now fairly substantial [1.3, 1.4]. Overall, the decay data of those radionuclides are well-known, except for positron emission branches in a few cases [1.5] (see also IAEA report INDC(NDS)-0535 (2008)). A new IAEA-CRP has been proposed, and is expected to remove the deficiencies in these data, and lead to a standardised cross-section database for the production of these radionuclides.

As far as internal radiotherapy is concerned, enhanced interest is manifested in radionuclides that emit low-energy Auger and conversion electrons or  $\alpha$  particles. Available information on decay data is often incomplete, while sophisticated interdisciplinary techniques, such as clean chemical separations and X-ray spectrometry or  $\alpha$ -counting, are needed to address difficulties in the production methods. As an example, recent work was described on the production of the Auger electron emitter  $^{193}\text{Pt}^{\text{m}}$  with high specific activity via the  $^{192}\text{Os}(\alpha,3\text{n})$  process [1.6].

Nuclear data work at Jülich relevant to hadron therapy was limited to the determination of the activation products in human tissue and collimator materials. The measured data for the  $^{\text{nat}}\text{N}(\text{p},\text{x})^{11}\text{C}$ ,  $^{\text{nat}}\text{N}(\text{p},\text{x})^{13}\text{N}$  and  $^{\text{nat}}\text{O}(\text{p},\text{x})^{13}\text{N}$  reactions up to  $\sim 200$  MeV in collaboration with PSI and iThemba Laboratory were used to calculate the total activity in the brain generated during proton therapy. The extra dose caused by the activation products in relation to the dose during therapy was quantitatively shown for the first time to be negligible [1.7]. Several soft radiation emitting activation products in the collimator materials were also determined [1.8].

As far as future data needs are concerned, the development of novel positron emitters and therapeutic radionuclides will keep the community engaged for many years, particularly if full information is needed on the formation of low-energy electrons in the decay of therapeutic radionuclides. The use of intermediate energy reactions and multi-particle beams ( $^3\text{He}$ ,  $\alpha$ , etc.) for the production of radionuclides may be advantageous, but will call upon more nuclear data efforts. Further measurements of activation products in proton as well as heavy-ion therapy may also be required.

- [1.1] S.M. Qaim, Nuclear data relevant to cyclotron produced short-lived medical radioisotopes, *Radiochim. Acta* **30** (1982) 147-162.
- [1.2] S.M. Qaim (Editor), *Nuclear Data for Medical Applications*, Special issue of *Radiochim. Acta* **89** (2001) 189-355.
- [1.3] S.M. Qaim, Decay data and production yields of some non-standard positron emitters used in positron emission tomography, *Quart. J. Nucl. Med. Mol. Imaging* **52** (2008) 111-120.

- [1.4] S.M. Qaim, Development of novel positron emitters for medical applications: nuclear and radiochemical aspects, *Radiochim. Acta* **99**, 611-625 (2011).
- [1.5] S.M. Qaim, *et al.*, Positron emission intensities in the decay of  $^{64}\text{Cu}$ ,  $^{76}\text{Br}$  and  $^{124}\text{I}$ , *Radiochim. Acta* **95** (2007) 67-73.
- [1.6] M.S. Uddin, *et al.*, Radiochemical determination of cross sections of alpha particle induced reactions on  $^{192}\text{Os}$  for the production of the therapeutic radionuclide  $^{193}\text{Pt}$ , *Appl. Radiat. Isot.* **68** (2010) 2001-2006.
- [1.7] K. Kettern, *et al.*, Quantification of radiation dose from short-lived positron emitters formed in human tissue under proton therapy conditions, *Radiat. Phys. Chem.* **78** (2009) 380-385.
- [1.8] S.M. Qaim, *et al.*, Excitation functions of nuclear reactions leading to the soft-radiation emitting radionuclides  $^{49}\text{Ca}$ ,  $^{49}\text{V}$  and  $^{204}\text{Tl}$  in beam collimator materials used in proton therapy, *Radiochim. Acta* **98** (2010) 447-457.

## 2.2. Medical Applications Over the Intermediate Term: Decay Data Requirements, A.L. Nichols (University of Surrey, UK)

As an evaluator and assembler of sets of recommended decay data for actinides and their heavy-element decay products, activation products, fission products and standards/calibrants, A.L. Nichols focused on the requirements and availability of atomic and nuclear decay data in support of nuclear medicine applications. Staff of the IAEA Nuclear Data Section provide rapid and convenient access to a significant number of such data sets, based on a strategic intent to create relational databases, and Web and CD multiplatform communication processes, with regular and sometimes continuous updating of specific sets of data that are applicable to nuclear medicine and analytical science, as well as nuclear power generation.

Consideration has been given to databases that are judged to be most relevant to decay-data needs in nuclear medicine:

- (1) ENSDF (Evaluated Nuclear Structure Data File) – nuclear structure and decay data for all known radionuclides ([www.nndc.bnl.gov/ensdf/](http://www.nndc.bnl.gov/ensdf/)) Evaluated at regular intervals (normally every seven to ten years) by members of the International Network of Nuclear Structure and Decay Data Evaluators, and published in *Nuclear Data Sheets*. The strength of ENSDF is to be found within the completeness of this comprehensive database.

The NuDat electronic chart of the nuclides constitutes a user-friendly means of extracting decay data from ENSDF ([www.nndc.bnl.gov/nudat2/](http://www.nndc.bnl.gov/nudat2/)); while MIRD (Medical Internal Radiation Dose) generates the result of processing ENSDF decay data through RADLST to give the energies and intensities of all emitted radiation, along with their dose rates ([www-nds.iaea.org/mird/](http://www-nds.iaea.org/mird/)). NSR (Nuclear Science References) is an ancillary bibliographic database upon which ENSDF is based ([www-nds.iaea.org/nsr/index.jsp](http://www-nds.iaea.org/nsr/index.jsp)).

- (2) DDEP (Decay Data Evaluation Project) – comprehensive atomic and nuclear decay data evaluations of **selected** radionuclides, with significant emphasis placed on the completeness of each decay scheme and the derivation of X-ray and electron decay data. Multinational evaluations are submitted via the coordinator to the custodian of the database and comments file at the Laboratoire National Henri Becquerel, CEA Saclay, France ([www.nucleide.org/DDEP\\_WG/DDEPdata.htm](http://www.nucleide.org/DDEP_WG/DDEPdata.htm)).

Decay data for 190 radionuclides are contained within DDEP (on 4 August 2011), of which the following are directly applicable in nuclear medicine:  $^{11}\text{C}$ ,  $^{13}\text{N}$ ,  $^{15}\text{O}$ ,  $^{18}\text{F}$ ,  $^{32}\text{P}$ ,  $^{60}\text{Co}$ ,  $^{64}\text{Cu}$ ,  $^{66}\text{Ga}$ ,  $^{67}\text{Ga}$ ,  $^{89}\text{Sr}$ ,  $^{90}\text{Y}$ ,  $^{99\text{m}}\text{Tc}$ ,  $^{111}\text{In}$ ,  $^{123}\text{I}$ ,  $^{125}\text{I}$ ,  $^{131}\text{I}$ ,  $^{137}\text{Cs}$ ,  $^{153}\text{Sm}$ ,  $^{159}\text{Gd}$ ,  $^{166}\text{Ho}$ ,  $^{169}\text{Yb}$ ,  $^{177}\text{Lu}$ ,  $^{186}\text{Re}$ ,  $^{188}\text{Re}$ ,  $^{192}\text{Ir}$ ,  $^{201}\text{Tl}$ ,  $^{211}\text{At}/^{211}\text{Po}$  and  $^{213}\text{Bi}/^{225}\text{Ac}$ . Amongst others,

serious consideration needs to be given to the future inclusion of  $^{45}\text{Ti}$ ,  $^{52}\text{Mn}$ ,  $^{52}\text{Fe}$ ,  $^{55}\text{Co}$ ,  $^{61}\text{Cu}$ ,  $^{62}\text{Cu}$ ,  $^{67}\text{Cu}$ ,  $^{72}\text{As}$ ,  $^{73}\text{Se}$ ,  $^{75}\text{Br}$ ,  $^{76}\text{Br}$ ,  $^{77}\text{Br}$ ,  $^{82}\text{Rb}$ ,  $^{82}\text{Sr}$ ,  $^{83}\text{Sr}$ ,  $^{86}\text{Y}$ ,  $^{90}\text{Nb}$ ,  $^{94}\text{Tc}^{\text{m}}$ ,  $^{103}\text{Pd}$ ,  $^{110}\text{In}^{\text{m}}$ ,  $^{120}\text{I}$ ,  $^{121}\text{I}$ ,  $^{124}\text{I}$ ,  $^{131}\text{Cs}$ ,  $^{140}\text{Nd}$ ,  $^{165}\text{Er}$  and  $^{193}\text{Pt}^{\text{m}}$ .

- (3) Relevant IAEA Coordinated Research Projects and databases –  
Evaluated X-ray and  $\gamma$ -ray standards (1998-2005)

[www-nds.iaea.org/xgamma\\_standards/](http://www-nds.iaea.org/xgamma_standards/)

[www-nds.iaea.org/publications/tecdocs/sti-pub-1287\\_Vol1.pdf](http://www-nds.iaea.org/publications/tecdocs/sti-pub-1287_Vol1.pdf)

[www-nds.iaea.org/publications/tecdocs/sti-pub-1287\\_Vol2.pdf](http://www-nds.iaea.org/publications/tecdocs/sti-pub-1287_Vol2.pdf)

Nuclides within this particular database include  $^{60}\text{Co}$ ,  $^{64}\text{Cu}$ ,  $^{66}\text{Ga}$ ,  $^{67}\text{Ga}$ ,  $^{99}\text{Tc}^{\text{m}}$ ,  $^{111}\text{In}$ ,  $^{123}\text{I}$ ,  $^{125}\text{I}$ ,  $^{131}\text{I}$ ,  $^{137}\text{Cs}$ ,  $^{153}\text{Sm}$ ,  $^{166}\text{Ho}$ ,  $^{169}\text{Yb}$ ,  $^{192}\text{Ir}$  and  $^{201}\text{Tl}$ .

Updated actinide decay data library (2005-2011) – data forwarded to DDEP

[www.nucleide.org/DDEP\\_WG/DDEPdata.htm](http://www.nucleide.org/DDEP_WG/DDEPdata.htm)

Nuclides within this particular database include  $^{212}\text{Bi}$  ( $^{228}\text{Th}$  decay chain),  $^{213}\text{Bi}$  ( $^{229}\text{Th}/^{225}\text{Ac}$  decay chain),  $^{211}\text{At}/^{211}\text{Po}$ ,  $^{223}\text{Ra}$  (within  $^{227}\text{Ac}$  decay chain),  $^{241}\text{Am}$  and  $^{252}\text{Cf}$ .

New work is required to build on these previous IAEA CRPs through consideration of possible nuclear data needs over the next 5 to 15 years (from 2011 to 2025). Improvements to the existing data are required, along with radionuclidic additions to ensure that some pre-emptive measures are in place to address future needs. For example, the development of metallic-based positron emitters can be envisaged as a consequence of recent improvements in their separation chemistry (Ti and Cu radionuclides), while the advent of microdosimetry may underline the need for suitable low-energy Auger-electron emitters for therapeutic applications.

### 2.3. International Links and Relevant Activities at Laboratoire National Henri Becquerel (LNHB), M.-M. Bé (CEA/LNHB, France)

The existing international radionuclide metrology chain was described, from the Bureau International des Poids et Mesures (BIPM) at the top to the end data users (for example, hospital practitioners).  $^{18}\text{F}$  was taken as an example - the first measurement of activity is carried out by means of a ‘primary method’ which does not require calibration - the result of this activity measurement is taken as the reference value, which is transferred as a ‘point of calibration’ to secondary instruments such as an ionization chamber (IC). When users wish to calibrate their own instrument (such as radiometers to measure activity in hospitals), they send a sample of  $^{18}\text{F}$  solution to the national metrology institute (NMI). IC activity measurements are carried out on this sample, and the results are transmitted to the user, along with an official ‘certificate’. Standards are needed at each level of this measurement chain.

The Laboratoire National Henri Becquerel (LNHB) is the French NMI, and makes use of various specific instruments to maintain and upgrade the references for activity (Becquerel) and dosimetry (Gray). Since all methods require a good knowledge of the decay data, a small group in LNHB is responsible for the determination of recommended data. LNHB was also one of the creators of the DDEP international working group (Decay Data Evaluation Project), with the objective of producing well-defined decay data that can be used by the ionizing radiation metrology community and by all the users of ionizing radiations.

The evaluations done by the DDEP members, when available, are sent to LNHB for publication. LNHB/DDEP was given the opportunity to publish the results of the DDEP evaluations as a BIPM Monographie, under the auspices of the CCRI (Consultative Committee for Ionizing Radiations). A total of six volumes have now been published, and a web site of the data files ([www.nucleide.org](http://www.nucleide.org)) is regularly updated.

The second part of the presentation was dedicated to possible ‘emerging radionuclides’. A list of novel radionuclides suggested by medical physicists was shown. However, other nuclides are considered as important elsewhere, and the list of potentially useful radionuclides can be very long. Therefore, a first action should be to select a restricted list of nuclides judged important and common to several countries. Depending on the route of production, some impurities present in the final solution also need to be determined in a quantitative manner. M.-M. Bé highlighted during her presentation that the decay scheme data are not well-known for most of these nuclides and impurities - a reference value in the international system of references (SIR) does not exist, and therefore traceability cannot be assured for these radionuclides.

This situation will be slightly improved in the near future since two international exercises have been carried out:

- (1) Activity measurements on a solution of  $^{177}\text{Lu}$  by twelve laboratories participating in a Key Comparison Reference Data project designed to create an international standard.
- (2) Five European NMIs have undertaken  $^{64}\text{Cu}$  activity measurements. They have been asked to determine the activity of a  $^{64}\text{Cu}$  solution using all available/possible measurement techniques, send a sample to the BIPM for submission to SIR, and measure the decay data. Results of five activity measurements have been sent to the SIR - good agreement has been observed, and therefore a key reference value will be established. Moreover, a new decay scheme will be derived from previously published decay data and the new results obtained in this exercise.

Some potential medical radionuclides are  $\beta^-$  emitters. Knowledge of the dose delivered within the tumour and the rest of the body is essential to ensure efficacy and safety in any treatment using ionizing radiations. Dose assessments require an accurate definition of the energy spectrum of the  $\beta^-$  transition, but the necessary data are not known for emerging nuclides with unmeasured beta spectra (spectra have to be calculated by means of theoretical models). While the shapes of the beta spectra depend on the nature of the transition (allowed, unique forbidden, non-unique forbidden), existing computer programs are only able to calculate the spectra for allowed and unique forbidden transitions. In order to improve the existing codes and extend such calculations to non-unique forbidden transitions, new measurements of  $\beta^-$  spectra are required. These studies will help derive experimental shape factors from which reductions of the theoretical calculations will be made possible.

The following list of actions was suggested:

- (1) prioritize the nuclides to be studied;
- (2) support and encourage the production of these nuclides in order to provide sufficient quantities to permit activity and decay-data measurements;
- (3) determine and study impurities;
- (4) support and encourage measurements of beta spectra for relevant nuclides; and
- (5) evaluate decay scheme data after completion of new measurements.

#### 2.4. Nuclear Data Needs for Medical Isotope Research, F.G. Kondev (Argonne National Laboratory, USA)

Brief descriptions were given of the Office of Nuclear Physics, Office of Science, US DOE, the Isotope Development of Production for Research and Applications (IDPRA) Program, and various experimental facilities in USA where isotope production and R&D activities are carried out. The primary USA source of isotopes for science, medicine, security and applications is the National Isotope Data Center, which manages the coordination of isotope production across the many DOE-funded facilities, as well as private commercial facilities for some of the more important isotopes.

Nuclear data capabilities for the study of medical isotopes at Argonne National Laboratory were described. The main emphasis was on the ATLAS facility at which beams of practically all stable isotopes can be accelerated at energies above the Coulomb barrier and used to produce a variety of isotopes. Description of some of the ANL state-of-the art detector equipment available for medical isotopes measurements was also presented.

Nuclear decay data include half-lives, radiation emission energies and probabilities, mixing ratios and conversion coefficients for gamma emissions, and the atomic data associated with radioactive decay. Although the available data on half-lives are generally believed to be in good shape, the half-life of  $^{186}\text{Re}^m$  was judged to be ill-defined. While several measurements may be available for the gamma-ray emission probabilities of a particular nuclide, greater attention should be paid in cases where direct decay between the parent and ground states of the daughter nuclide is involved. Usually dedicated studies are required to deduce the normalization factor between the relative and absolute emission probabilities. Examples were presented for  $^{67}\text{Ga}$  and  $^{67}\text{Cu}$ , in which uncertainties in the ground-state to ground-state emission probabilities can lead to more than 10% uncertainty in the measured cross sections and dose calculations. New measurements of  $^{67}\text{Cu}$  have been initiated at ANL in order to remove these ambiguities. Many existing decay-data evaluations (ENSDF and DDEP) possess a lack of information on the quality of the available and recommended data and the need for further improvements. Thus, such data assessments are required for the medical isotopes under consideration.

Decay data needs for impurity isotopes produced in various neutron and charged particle production reactions were discussed in details. While radiochemistry methods are usually used to achieve a high-purity of a particular medical radioisotope, sound knowledge of the decay data for the produced impurity nuclei is also important. A comprehensive study needs to be performed in order to identify the main impurity nuclides that can be produced within the main production reactions, coupled together with the available purity of the target materials, and to assess the corresponding decay data needs. An example was presented on the importance to quantify  $^{177}\text{Lu}^m$  ( $T_{1/2} = 160.44$  d) impurity in the production of the therapeutic nuclide  $^{177}\text{Lu}$ . Studies were recently performed at ANL, where a new decay branch from the longer-lived  $^{177}\text{Lu}^m$  isomer was discovered. New beta- and IT-decay branching intensities from the isomer were also deduced, which were found to differ by ~10% from the previously evaluated values.

Inadequacies were noted in the treatment of Auger, Coster-Kronig, super Coster-Kronig and other shake-off electrons following decay of radiotherapeutic Auger nuclides. Available computer programs and methodologies used to calculate the energy spectra and emission probabilities for such transitions are limited to only K- and L shells (EMISSION), and they also neglect the presence of secondary vacancies and use binding energies for neutral atoms. Experimental data for such processes are scarce. A new model aimed at improving the low-energy electron data is being developed at the Australian National University: results for  $^{55}\text{Fe}$

EC decay were presented in which good agreement for the KLL, KLX and KXY transitions was found between the new model and all other known approaches. The importance of the very low-energy transitions involving the M, N, O and other shells were demonstrated in the calculated energy spectra and emission probabilities for  $^{99}\text{Tc}^m$ . Collaboration between ANU and ANL continues to develop the new model further into a practical tool for the calculation and evaluation of energy spectra and emission probabilities for all Auger emitters.

Photonuclear reactions such as  $(\gamma, p)$  and photofission should be considered for the production of several isotopes of interest, particularly the approach being pursued at the TRIUMF facility to utilize photofission on  $^{238}\text{U}$  to produce  $^{99}\text{Mo}/^{99}\text{Tc}^m$ . The role of future radioactive beam facilities for the production of medical isotopes was discussed, such as the Facility for Rare Isotope Beams (FRIB) that is under construction at Michigan State University. An extensive range of radionuclides will be harvested, including some of importance in nuclear medicine, such as  $^{67}\text{Cu}$ ,  $^{44}\text{Ti}$ , etc.

## **2.5. Neutron Data for Proton and Carbon Beam Therapy, B.V. Carlson (Instituto Tecnológico de Aeronáutica (ITA), Brazil)**

Proton and carbon beam therapy have the advantage over photon therapy of being fairly precise in the localization of dose deposition within the region of the Bragg peak. The principal interactions are electronic and result in stopping; elastic Coulomb and nuclear reactions also contribute to stopping and spread of the beam. Non-elastic nuclear reactions deplete the beam and furnish reaction products and secondary particles that should be taken into account when analyzing the effects of radiation therapy.

The rate of beam depletion is determined by the total non-elastic cross sections of the beam particles with nuclei in the body or the delivery system. Cross sections important for dose calculations are the double differential cross sections for neutron and light-particle emission and photon and product recoil spectra. Activation cross sections of positron-emitting nuclei are of special interest, because their decay can be used to analyze beam localization. The elements of greatest interest are those most common in the body (H, C, O, N, P and Ca), and the structural elements of the delivery system (Si, Al, Fe, Cu, W and Pb).

The ideal method for calculating the effects of beam radiation therapy is with a Monte-Carlo transport code. Such calculations allow one to describe the source geometry with precision, take into account all physical interactions, and follow all secondary particles. These calculations require either an extremely complete cross-section database or reasonably detailed physical models and model parameters. A disadvantage of Monte-Carlo transport calculations is that they are normally time-consuming, although the increasing availability of parallel computing facilities are diminishing this drawback.

Experiments, as well as Monte-Carlo transport calculations, show that non-elastic nuclear reactions in the body reduce the dose contribution of a proton beam at the Bragg peak by about 5% for a 100-MeV beam, and by about 20% for a 200-MeV beam. The effect is much more severe for a carbon beam – over 2/3 of a 400A-MeV carbon beam is depleted before the Bragg peak, which occurs at about the same depth as a 200-MeV proton beam (at about 25 cm). Non-elastic proton-nucleus cross sections are well described by the Koning-Delaroche optical potential for nuclei heavier than aluminum, although transport codes normally use parameterizations that are less precise but extend to higher energies. No good proton optical model description is available for the light nuclei prevalent in the body (C, N and O), and clearly the parameterized cross sections furnish a better description of the data in these cases. Transport codes also use parameterizations of the carbon-induced non-elastic cross sections, which provide a reasonably precise description of the available data.

The most important activation products of radiation beam therapy are the relatively long-lived positron-emitters ( $^{11}\text{C}$ ,  $^{13}\text{N}$  and  $^{15}\text{O}$ , with half-lives of 20.3, 10.0 and 2.0 min, respectively);  $^{30}\text{P}$  and  $^{38}\text{K}$  positron-emitters, with half-lives of 2.5 and 7.6 min, also occur. A proton beam creates these products fairly uniformly over the full energy range, with production decreasing close to the Bragg peak where the beam no longer has sufficient energy for activation. The distribution is quite different for a carbon beam, in which the beam contributes to the activation products, furnishing a very large peak in the product distribution immediately before the Bragg peak.

A carbon beam produces a wide distribution of stable and unstable activation products along the path of the beam and beyond the Bragg peak, due to the importance of fragmentation reactions at the beam energies involved. The longest-ranged charged particles are the lower-charged isotopes of hydrogen and helium due to the slower stopping of these fragments. Magnitudes of the elemental fragment distributions can be estimated within a factor of approximately two.

Neutron production must also be taken into account in reactions induced by both proton and carbon beams, due to their long range. High-energy neutrons can produce secondary charged particles that deposit energy far from the intended irradiation region, while low-energy neutrons can be captured to produce residues that might decay later. Although the double differential distributions of emitted neutrons (and light particles in general) are poorly described by calculation, the total effect of their induced secondary reactions is estimated to be of the order of 1% of the total dose.

Ideally, planning of radiation beam treatment should be based on reliable calculations of the distribution of the dose to be deposited. Monte-Carlo transport calculations provide the most trustworthy means of achieving this aim by taking into account variations in beam delivery, tissue and bone density, and obstructions near the beam path. Such calculations require either extremely complete nuclear reaction data or validated nuclear models and parameter sets that describe the possible reactions with precision. The nuclear data most in need of improvement for proton beam therapy are as follows:

- (1) non-elastic cross sections of the light elements (C, N and O);
- (2) activation cross sections of residual nuclei, particularly the positron-emitters  $^{11}\text{C}$ ,  $^{13}\text{N}$ ,  $^{15}\text{O}$ ,  $^{30}\text{P}$  and  $^{38}\text{K}$ ; and
- (3) double differential emission cross sections of the light particles (particularly neutrons).

Due to the complexity of the fragmentation reactions that occur in a carbon beam, data sets that describe these reactions well would be extremely difficult to prepare. Therefore, the best course of action would appear to be the development of more precise models and validated parameter sets to describe the fragmentation and the production of light particles and residues. The derivation of better models and validated parameters would also be the ideal approach to proton beam therapy, in order to undertake more precise Monte-Carlo transport calculations of the effects on dose deposition of variations in morphology or in structure arising from bone or implants.

## **2.6. Some Thoughts on Data Integrity in Stacked-Foil Experiments, G.F. Steyn (iThemba Laboratory, South Africa)**

G.F. Steyn presented an overview on stacked-foil activation experiments and pointed out that these techniques not only found application in the measurement of excitation functions, but

also in the measurement of angular distributions and recoil range distributions of heavy target-like residues. These latter measurements proved to be very effective in identifying reaction mechanisms where a significant amount of momentum is carried away from the entrance channel by spectator fragments during a nuclear interaction (e.g., partial fusion and transfer reactions of complex projectiles with target nuclei). Such a phenomenon is especially important in  $^{12}\text{C}$ -induced reactions of relevance to ion beam therapy. Indeed, a theoretical approach developed to give a comprehensive description of break-up-fusion reactions based on a Generalized Serber Approximation in conjunction with the Boltzmann Master Equation (BME) for the subsequent pre-equilibrium decay has recently been incorporated into the FLUKA code. While stacked-foil activation experiments are conceptually quite simple, they should not be underestimated - they are not trivial and sometimes the saying that “the devil is in the detail” proves to be true.

The role of monitor excitation functions in stacked-foil experiments was considered in detail, particularly with respect to their role in the determination of the beam energy and accumulated charge. Although a number of new measurements have been proposed in the literature for this purpose, some of these values were determined relative to existing monitor reaction cross sections only. The measurement of monitor reaction cross sections requires a primary determination of the incident beam energy by means of either deflection in a calibrated magnetic field, or by time-of-flight (TOF) techniques, as well as a primary measurement of the integrated charge which requires the use of a calibrated Faraday cup or chamber with proper electron suppression as the target holder. Examples of relatively simple, small and inexpensive TOF apparatus and Faraday chambers were shown, as published in the literature - they can easily be established in the majority of available facilities. Furthermore, logging of the accumulated charge in small time intervals is useful to correct for beam intensity fluctuations during bombardment, which can improve the experiment in the case of short-lived radionuclides. Once established, these facilities can be useful for any stacked-foil activation experiments. However, such arrangements do not preclude the necessity for monitor reactions; on the contrary, multiple types of monitor foil should be interspersed throughout the stack to diagnose the consistency of the measured data. This approach will help to identify certain kinds of problems (e.g., an energy shift which may become more pronounced towards larger penetration depth within a stack).

Alternatives to the use of thin foils for determining excitation functions in activation experiments have been discussed, namely “the pellet methods”, and some results from such experiments were shown. While these methods are useful in a few special cases, they are not generally applicable and cannot replace the need for thin targets. The preparation of thin specimens of very reactive elements as well as of some brittle non-metallic elements can be problematic - more stable compounds are usually employed and several methods of target preparation have been published in the literature. Nevertheless, target damage during the bombardment is not unusual, and experimentalists should assess the integrity of their foils before and afterwards. While most of the recently published data seem to be of an acceptable quality, there are some notable exceptions, which is sometimes quite puzzling. The IAEA should consider publishing a set of guidelines to experimenters, particularly to aid newcomers to the field. New cross-section measurements, as foreseen in any future CRP, will have to be of sufficient quality to resolve existing discrepancies, and to identify and correct deficiencies in existing recommended data.



## 2.7. Possible Systematic Errors in Cross-section Measurements: Influence on Data Evaluations, S. Takács (Institute of Nuclear Research of the Hungarian Academy of Sciences, Debrecen, Hungary)

Systematic errors can be introduced at different stages of an experiment as well as during the data processing. S. Takács noted that during an irradiation, different systematic errors in the experimental measurements and data processing can be introduced to influence the final result. Systematic errors can also be introduced in data processing with the introduction of conversion factors, or applying improper correction and normalization factors to the experimental data.

The simplified equation used to deduce the cross section  $\sigma$  is given by Equation (1):

$$\sigma = \frac{T_{\gamma} \lambda}{\varepsilon_d \varepsilon_{\gamma} N_t N_b (1 - e^{-\lambda t_b}) e^{-\lambda t_c} (1 - e^{-\lambda t_m})} \quad (1)$$

where  $T_{\gamma}$  is the measured peak area,  $\varepsilon_d$  is the detector efficiency,  $\varepsilon_{\gamma}$  is the abundance of the gamma line investigated,  $N_t$  is the number of target nuclei,  $N_b$  is the number of bombarding particles,  $\lambda$  is the decay constant ( $\ln 2/T_{1/2}$ ), and  $t_b$ ,  $t_c$  and  $t_m$  are the irradiation time, cooling time and measuring time, respectively.

Possible systematic errors are considered below.

Peak area: should be corrected for different effects such as dead time when measuring at high count rate, summing effect using large volume detector and short detector-source distances, self-absorption for low-energy gammas and relatively thick target foils, random coincidence, different type of interferences and recoil effect, etc. Peak area is an experiment-specific parameter that is not published - therefore difficult to correct.

Detector efficiency: should be precisely determined, especially in the low-energy region used in the assessment of medical radioisotopes (where the log-log curve is not linear anymore). Such efficiency curves are specific to particular detectors, and are normally not published.

Gamma branching ratio: only parameter that contributes linearly to the uncertainty – values and uncertainties are known from data libraries, and therefore their effect can be corrected at a later stage if necessary. This parameter has usually been published.

Number of target atoms: can be determined by different methods - direct thickness measurements, mass and surface measurements for very thin targets, etc. Should consider checking for homogeneity and surface roughness, possible cracks, and pin holes. If a correction for target thickness is required, changes in the energy scale will also occur in the stacked target technique. Experiment specific.

Number of bombarding particles: determined from collected charge, or beam current measurement – specific to the experiment, and normally published. A time-averaged mean value is determined for pulsed and bunched beams. The beam intensity should be kept constant during the irradiation. If the beam parameters are determined by means of monitor reactions, a series of monitor foils with more than one reaction is advisable.

The above parameters contribute linearly, and their systematic errors can be corrected.

Time parameters: irradiation time, cooling time and measuring time do not contribute linearly. Only the irradiation time is published, while the cooling and measuring times are normally not given. Corrections are not easy, but possible systematic errors involving the half-life and time parameters of medical radionuclides are minimal since their values are well measured.

Decay constant or half-life: half-lives are taken from evaluated data libraries. Since this parameter does not contribute linearly, the effect of the systematic uncertainty in the time-based information cannot be properly corrected. Irradiation and cooling times can influence the deduced cross section: 5% systematic uncertainty in the half-life and a long cooling time (more than ten half-lives) can result in 30% uncertainty in the cross section. The systematic uncertainty in a short half-life can influence the shape of the measured excitation function, or result in scattered experimental cross-section data. Ratio of the excitation functions for two reactions can also be distorted.

During an evaluation process in which the experimental conditions and resulting data sets are considered, only the gamma abundances can be properly corrected. Other parameters are specific to the particular experiment, and are either not or only partially published – therefore, sound and confident correction of the systematic uncertainties is not possible.

## 2.8. Measurement and Use of Nuclear Data for Medical Radionuclides at the Nuclear Physics Institute, Academy of Sciences of the Czech Republic, O. Lebeda (Czech Academy of Sciences, Czech Republic)

### Measurement of excitation functions:

- $^{75}\text{As}(^3\text{He},2n)^{76}\text{Br}$  reaction (at the time of measurement,  $^{76}\text{Br}$  with half-life of 16.2 h was one of the most interesting non-conventional positron emitters; although this route requires  $^3\text{He}$  beam, use can be made of a naturally monoisotopic element ( $^{75}\text{As}$ )).
- $^{231}\text{Pa}(p,2n)$  and  $^{231}\text{Pa}(d,3n)$  reactions to estimate their production potential for alpha-emitting  $^{230}\text{U}/^{226}\text{Th}$  (20.8 d/30 min) and provide missing experimental data; found that the yield is low due to significant occurrence of fission, and thus similar to that of the indirect production via the  $^{232}\text{Th}(p,3n)^{230}\text{Pa}$  (17.4 d,  $\beta^-$  of 7.8 %)  $\rightarrow$   $^{230}\text{U}$ .
- $^{\text{nat}}\text{Mo}(p,x)$  and  $^{\text{nat}}\text{Mo}(d,x)$  reactions with regard to the formation of  $^{95}\text{Tc}^{\text{m}}$ ,  $^{96}\text{Tc}^{\text{g+m}}$ ,  $^{99}\text{Tc}^{\text{m}}$  and  $^{99}\text{Mo}$ . Proton-induced reactions leading to  $^{99}\text{Tc}^{\text{m}}$  and  $^{99}\text{Mo}$  take place almost entirely on  $^{100}\text{Mo}$ , and therefore the measured data could be converted to isotopic cross sections for the formation of these medically relevant radionuclides. The measured yields of  $^{99}\text{Tc}^{\text{m}}$  confirm the possibility of cyclotron-based production of this important radionuclide at the level of 1 TBq (at delivery time). There are several serious questions to be solved with respect to this alternative approach to  $^{99}\text{Tc}^{\text{m}}$  production (see below).
- $^{\text{nat}}\text{Nd}(p,x)$  reactions for estimating possible activation of freshly mined Nd by cosmic protons during transport for use in the SNO+ experiment, and also to provide missing data.

### Production of radionuclides:

- Design, testing and routine operation of gaseous target systems for production of  $^{81}\text{Rb}$ ,  $^{83}\text{Rb}$  and  $^{123}\text{I}$ .
- Design, testing and routine operation of the liquid target system for production of  $^{18}\text{F}$ .
- Design and testing of solution-based target systems for the production of  $^{86}\text{Y}$  and  $^{68}\text{Ga}$ .
- Design, testing and operation of solid target systems for production of  $^{124}\text{I}$  (external) and  $^{208}\text{Po}$ ,  $^{211}\text{At}$  (internal).
- Design and testing of solid target systems for production of  $^{61,64,67}\text{Cu}$  (both external and internal), and  $^{230}\text{U}$  via  $^{232}\text{Th}(p,3n)$  reaction (external).
- Implemented, but currently not used for production of  $^{67}\text{Ga}$ ,  $^{111}\text{In}$  and  $^{201}\text{Tl}$ .
- Interest in non-conventional diagnostic and therapeutic radionuclides, e.g.,  $^{43,44,47}\text{Sc}$  and Auger emitters.

### Recommended nuclear data measurements:

When choosing new radionuclides for nuclear data measurements (both production routes and decay data), the following priorities should be implemented.

Gamma emitters: suitable half-life of the order of hours to tens of hours, one or two gamma lines between 100 to 250 keV with sufficiently high intensity, and no or low particle emission accompanying the decay (preferably EC- or IT-decaying radionuclides); acceptable labelling chemistry, with no significantly radiotoxic decay products

Positron emitters: suitable half-life of the order of hours to tens of hours, high probability of positron emission of low energy, and no or low emission of gammas accompanying the decay (particularly those close to or of higher energy than the 511-keV annihilation peak); acceptable labelling chemistry, and no significantly radiotoxic decay products

Therapeutic radionuclides: suitable half-life of the order of a few days, high probability of electron emission (energies might vary, since therapy requirements vary according to the type of tumour – the more extensive the energy region covered, the better; however, recent interest has focused on low-energy beta emitters), emission of gammas or positrons (e.g.,  $^{64}\text{Cu}$ ) suitable for imaging of the biodistribution of the therapeutic dose is highly desirable;

alpha emitters - both high- and low-energy alpha particles are of interest (maximum range of the most energetic alpha particles in tissue is no more than a few cell diameters, i.e., ca 0.1 mm); particular attention must be paid to high stability of the labelled *in vivo* compounds (chelated metal radionuclides such as  $^{213}\text{Bi}$ ,  $^{230}\text{U}/^{226}\text{Th}$  seem to be more promising in comparison to  $^{211}\text{At}$ , with favourable decay properties, but inadequate knowledge of chemical properties, and lower stability of compounds), no significantly radiotoxic decay products, if they cannot be used for therapy as well (e.g., very short-lived decay products of  $^{226}\text{Th}$  only enhance the curing effect of the mother radionuclide since they emit other alpha particles, but because of their short half-lives do not succeed to migrate from their place of origin).

### Suggested topics:

- a) Re-measurement of the decay data of  $^{43,44,47}\text{Sc}$ ,  $^{61,64,67}\text{Cu}$ , and  $^{230}\text{U}/^{226}\text{Th}$  (and decay chain to  $^{210}\text{Pb}$ ).
- b) Detailed study of the possible cyclotron production of  $^{99}\text{Tc}^{\text{m}}$ , including recommended cross-sections for the  $^{100}\text{Mo}(p,2n)$  reaction, measurement of the radionuclidic impurities produced in the target both by activation of  $^{100}\text{Mo}$  and residual stable Mo isotopes in enriched  $^{100}\text{Mo}$  (particular attention should be paid to Tc radioisotopes that cannot be chemically separated from the product); estimate the content of  $^{99}\text{Tc}^{\text{g}}$  in the  $^{99}\text{Tc}^{\text{m}}$  produced, and possible effect on labelling of commercially-available kits.
- c) Detailed measurement of the production routes for  $^{61}\text{Cu}$  with respect to optimal radionuclidic purity.
- d) New measurements of the somewhat scattered data of monitoring reactions for  $^3\text{He}$  on Al, Ti and possibly another suitable metal in order to improve the reliability of the currently recommended cross sections
- e) Precise calculations of the Auger, Coster-Kronig and super Coster-Kronig electron energies and emission probabilities for promising therapeutic radionuclides.

## **3. DISCUSSIONS**

Various needs for atomic and nuclear data will arise during the course of the next 5 to 15 years with respect to the development, followed by the adoption or rejection of particular radionuclides for diagnostic and therapeutic applications. Specific radionuclides were identified for consideration within the agreed timeframe, and discussions ensued as to the nature of the data requirements in the preparation of the desired high-purity nuclides with well-defined decay characteristics. A number of proposals and possibilities were considered,

and statements on requirements and their consequences are given below. Detailed radionuclidic assessments and requirements can be found in tabulated form within Section 4.

### **3.1. Cross-section Production Data, G.F. Steyn (iThemba Laboratory) and S. Takács (INR)**

Guidelines were given in two previous IAEA CRPs regarding the measurement of reaction cross sections<sup>1,2</sup>. Nevertheless, additional advice and further recommendations were formulated for stacked-foil activation experiments that are commonly used to determine the excitation functions of the nuclear reactions of interest.

#### Monitor reaction excitation functions:

- The beam energy should always be measured.  
This measurement may be performed by means of either magnetic deflection in a calibrated analysis magnet or time-of-flight (TOF) techniques. While analysis magnets may be too large for smaller facilities, compact and, inexpensive but accurate TOF techniques have been published in the literature, and should be within the financial reach of most laboratories in the field.
- The accumulated charge should always be measured.  
The target holder should be a Faraday cup or chamber, and there should be proper electron suppression.
- Use well-established monitor excitation functions for consistency checks, preferably more than one type of foil in the same experiment.
- Complete sets of monitor foils should be interspersed throughout the stack.  
Avoid, for example, using only one monitor foil as the first foil of the stack (which has been noted to occur in some publications).
- Use very high quality and well-characterized foils.  
Check the foils for damage after bombardment, especially in the case of thin, non-metallic samples.

#### Important aspects of stacked-foil experiments:

- Employ as many of the above features as feasible.
- Use of well-established monitor excitation functions for consistency checks (and preferably more than one type of foil in the same experiment) is highly recommended - provides a sensitive diagnostic to the overall quality of the data. For example, such a recommended approach would highlight an energy shift, especially towards deeper penetration depth in a target stack.
- Check the thickness and quality of each target foil individually - do not rely on the quoted value of the thickness given by the provider. Check the stability of the target foil during and after irradiation to prevent false results and contamination of the irradiation and measurement set-up.
- Too many foils in one stack can result in significant errors in the calculated energy scale, and may also influence the cooling and measuring times. Instead of one long stack, prepare two independent stacks with an overlapping energy region.
- If possible, use absolute beam energy and intensity measurements, and adopt only the monitor reactions as controls.
- If the beam parameters are determined from the monitor reaction, a series of monitor foils with more than one reaction is advisable.
- Use enriched targets and/or chemical separation to avoid serious interferences.
- Avoid cooling times that are long (i.e., no more than five half-lives).

- Apply total dead time correction (electronics + analyzer) and check for other possible corrections such as recoil effect, interferences, self-absorption, effect of summing and escape peaks, etc.
- Recoil effect can be high for a thin target and heavy, high-energy bombarding particles.
- Use the same well-defined measuring geometry for detector calibration and target foil measurements.
- Use the most up to date decay data for data processing.
- When preparing publications, always include lists that quantify the corrections applied during data processing.
- Give both the measured and estimated partial uncertainty components.
- List only those gamma lines and their branching ratios that were used in data processing.

### 3.2. Decay Data

A rigorous assessment of the available decay-data evaluations included in the DDEP and ENDSF databases is recommended to identify gaps in the experimental data and/or the decay properties adopted by the evaluator(s). Following such a comprehensive assessment, recommendations can be made for the adoption of an existing evaluation or initiation of a re-evaluation, as well as requests for new measurements if necessary. This all-encompassing requirement applies to all of the radionuclides tabulated in Section 4.

#### 3.2.1. Weak Gamma-ray Emissions

Specific weak gammas are sometimes used in particular cross-section studies:

1.  $^{64}\text{Cu}$  (recently re-evaluated for DDEP);
2.  $^{103}\text{Pd}$  (priority 1 evaluation within a planned CRP designed to address “improvements in charged-particle monitor reactions and nuclear data for medical radionuclides”, INDC(NDS)-0591, 2011);
3.  $^{211}\text{At}$  (re-evaluated within CRP on “Updated Actinide Decay Data Library, and placed on DDEP Web site).

Requirements for such well-defined, low-intensity decay data should always be born in mind during any research and development phase.

#### 3.2.2. Auger-electron Data, M.-M. Bé (CEA/LNHB)

A journal article entitled “The Amazing World of Auger Electrons” was recently published (A.I. Kassis, *Int. J. Radiat. Biol.* **80**, 11 (2004) 789), in which the author gives a summary of the investigations carried out over the previous 40 years to use Auger-electron emitting nuclides for medical purposes. The extreme radiotoxicity of low-energy electrons is attributed to the highly localized energy depositions in the immediate vicinity of the decay site (as implanted near the tumour to be destroyed), and has also encouraged scientists to investigate extensively their radio-biological effects at the molecular and cellular levels. Estimates of the absorbed dose in microscopic volumes require a detailed knowledge of Auger-electron spectra, including very low energy N- and O-shell Auger electrons and Coster-Kronig transitions. While a significant number of papers have been published on this subject over the past 20 years, the accuracy of the calculated Auger electron spectra depends on the availability of well-defined atomic and nuclear physics data.

(a) Activity measurements by means of Liquid Scintillation Counting (LSC) techniques

Although activity measurements of radionuclides decaying by electron capture transitions are usually carried out by adopting LSC, this technique is based on theoretical models that describe the interactions between the incident particles and the scintillator. Since electron capture causes a vacancy in the shells of the daughter nuclide, an electron rearrangement process follows in which Auger electrons or X-ray photons are emitted and interact with the atoms of the scintillator. This interaction is usually calculated with the aid of Monte-Carlo simulation codes, which require good knowledge of the energies and relative probabilities of the Auger electrons and X-ray photons.

The sequence of events must be described precisely:  $K_{\alpha}$  photon +  $L_i M_i M_j$  electrons, or KLL electrons + LMM electrons, etc. All possible paths are calculated in order to determine the effective energy deposited in the sample. Such a description requires knowledge of the K and  $L_i$  fluorescence yields, Coster-Kronig coefficients, and all the relative emission probabilities for the X-rays and Auger electrons (for more details see R. Broda, *et al.*, *Metrologia* **44** (2007) S36-S52).

If such nuclides are used for medical treatment, a good measurement of the activity of the solution delivered to the patient will be required, as underlined by Howell (*Int. J. Radiat. Biol.* **84**, 12 (2008) 959).

The following atomic parameters are required:

- binding electron energies;
- fluorescence yields,  $\omega_K$ ,  $\omega_{L1}$ ,  $\omega_{L2}$ ,  $\omega_{L3}$ ,  $\omega_{M_i}$ ,  $\omega_N$ , etc.;
- $\eta_{K L_1}$ ,  $\eta_{K L_2}$ ,  $\eta_{K L_3}$ , the number of vacancies created in the  $L_i$  sub-shell after vacancy transfers, when one K vacancy is filled, and similarly for the other  $M_i$ ,  $N_j$ , ... sub-shells;
- relative emission probabilities of the various possible X-ray emissions when a vacancy is created in K-,  $L_i$ -,  $M_i$ -,  $N_i$ -, ... shells, (*e.g.*, K- $L_2$ , K- $M_3$ , ...  $L_i$ - $N_2$ ,  $L_i$ - $M_3$ ,  $L_i$ - $O_{2,3}$ , ... );
- similarly for Auger electrons, K-, L-, M-, N- ... Auger-electron relative emission probabilities when a vacancy is created in K-,  $L_i$ -,  $M_i$ -,  $N_i$ -, ... shells, (*e.g.*,  $L_i$ -  $L_j M_k$ , etc.  $M_i$ -  $M_j O_k$ , ... );
- Coster-Kronig probabilities  $f_{12}$ ,  $f_{13}$ ,  $f_{23}$ , where  $f_{ij}$  is the number of vacancies created in the  $L_j$  sub-shell when an initial vacancy in the  $L_i$  sub-shell is filled by an electron of the  $L_j$  sub-shell, and similarly for the  $M_i$ ,  $N_i$ , ... sub-shells.

(b) Recommended actions

List existing publications and tables to determine if all parameters listed above are available or if some are missing.

Assess the quality of the existing publications and tables.

If necessary, recommend further studies to improve these parameters.

Encourage studies to fulfil the needs of the users for a more comprehensive and detailed calculation of Auger electrons.

### 3.3. Other Issues

#### 3.3.1. Isomeric States

Most of the commonly used diagnostic and therapeutic radionuclides do not have isomeric states (e.g.,  $^{11}\text{C}$ ,  $^{18}\text{F}$ ,  $^{90}\text{Y}$ ,  $^{123,125,131}\text{I}$  and  $^{201}\text{Tl}$ ). However, many metallic radionuclides do possess isomeric states with well-defined half-lives, as do some of the novel positron emitters and X-ray and low-energy electron-emitting therapeutic radionuclides. An isomeric impurity may distort the imaging quality of positron emitters, and also cause extra radiation dose to the patient; examples include  $^{73}\text{Se}^{\text{m,g}}$ ,  $^{86}\text{Y}^{\text{m,g}}$  and  $^{94}\text{Tc}^{\text{m,g}}$ . On the other hand, the existence of an isomeric state within a therapeutic radionuclide may constitute an ideal source of radioactivity – such a highly-converted internal transition leads to the emission of Auger electrons and X-rays (e.g.,  $^{193}\text{Pt}^{\text{m}}$  and  $^{195}\text{Pt}^{\text{m}}$ ). Hence, fundamental studies on isomeric states are of considerable value in the development of new radionuclides for medical applications.

Systematic studies near the thresholds of reactions have shown that the isomeric cross-section ratio depends on the spins of the two states concerned, and not on their energy of separation. Furthermore, thorough investigations have been undertaken of several isomer pairs, and the effects of different combinations of target, projectile, ejectile and input nuclear structure (S. Sudár and S.M. Qaim, *Phys. Rev.* **C53** (1996) 2885; K. Hilgers, *et al.*, *Phys. Rev.* **C76** (2007) 064601; M.S. Uddin, *et al.*, *Phys. Rev.* **C84** (2011) 024605).

The problem of long-lived isomeric states in medically interesting shorter-lived radionuclides was also discussed. Four important examples of this particular situation are  $^{99}\text{Tc}^{\text{m,g}}$ ,  $^{177}\text{Lu}^{\text{m,g}}$ ,  $^{186}\text{Re}^{\text{m,g}}$  and  $^{192}\text{Ir}^{\text{m,g}}$ :

$^{99}\text{Tc}^{\text{m,g}}$  has been well studied –  $^{99}\text{Tc}^{\text{m}}$  ( $T_{1/2} = 6.0$  h) is the isotope of interest, and the long-lived  $^{99}\text{Tc}^{\text{g}}$  ( $T_{1/2} = 2.1 \times 10^5$  y) is the isomer of concern;

$^{177}\text{Lu}^{\text{m,g}}$  – the ground state is of medical interest ( $T_{1/2} = 6.65$  d), while the metastable state constitutes the activity of concern;

$^{186}\text{Re}^{\text{m,g}}$  – the ground state is shorter lived ( $T_{1/2} = 89.3$  h) and of therapeutic interest, while the metastable state has a very long half-life of  $2.0 \times 10^5$  years;

$^{192}\text{Ir}^{\text{m,g}}$  – the shorter lived ground state ( $T_{1/2} = 73.8$  d) is used in radiotherapy, and the longer-lived metastable state is an undesired impurity ( $T_{1/2} = 241$  y).

Investigations of the levels of longer-lived impurities in the above isomeric combinations have been published in the open literature (K. Hilgers, *et al.*, *Appl. Radiat. Isot.* **63** (2005) 93; M. Hussain, *et al.*, *Radiochim. Acta* **98** (2010) 385), but more detailed studies are required. Serious problems arise from the longer-lived  $^{177}\text{Lu}^{\text{m}}$  in the case of  $^{177}\text{Lu}^{\text{g}}$ . However, the difficulty with the other three systems listed above is simply the presence of extra nuclei which decrease the specific activities of the desired medical products.

Improvements are required in both the cross-section and decay data for long-lived isomers, particularly for the cyclotron production of  $^{99}\text{Tc}^{\text{m}}$  which requires the equivalent production data for  $^{99}\text{Tc}^{\text{g}}$ . Similar detailed studies would also be beneficial for  $^{186}\text{Re}^{\text{m}}$  and  $^{177}\text{Lu}^{\text{m}}$ . Data such as the energy-dependent spin cut-off parameter, level densities at high excitation energies, and collective levels above the isomeric states need to be determined. Furthermore, the cross-section codes need to be improved over the intermediate energy range.

### 3.3.2. Cyclotron Production of $^{99m}\text{Tc}$

Over recent years, the supply of fission-produced  $^{99}\text{Mo}$ - $^{99m}\text{Tc}$  worldwide has become insecure. Therefore, much discussion has occurred as to the possibility of exploiting alternative routes of production of  $^{99m}\text{Tc}$ . The most promising means of addressing local/regional demands would appear to be the  $^{100}\text{Mo}(p,2n)$  reaction on highly-enriched  $^{100}\text{Mo}$  targets, covering a proton energy range of  $E_p = 22$  to  $12$  MeV (B. Scholten, *et al.*, Appl. Radiat. Isot. **51** (1999) 69). As far as improved nuclear data are concerned, a large number of relevant publications have recently appeared. Under these circumstances, available data for the  $^{100}\text{Mo}(p,2n)^{99m}\text{Tc}$  and  $^{100}\text{Mo}(p,pn)^{99}\text{Mo}$  reactions should be evaluated with high priority as part of the proposed CRP described in Ref. 4.

Detailed consideration was given to a specific list of reactions in connection with the cyclotron production of  $^{99}\text{Mo}$  ( $^{99m}\text{Tc}$ ), as provided by the Industrial Applications and Chemistry Section (IACS) of the IAEA (see Annex A). Both the  $^{100}\text{Mo}(p,p2n)^{98}\text{Mo}$  and  $^{100}\text{Mo}(p,p3n)^{97}\text{Mo}$  reactions generate stable products, and no cross-section measurements exist. Furthermore, the  $^{100}\text{Mo}(p,\alpha n)^{96}\text{Nb}$  and  $^{100}\text{Mo}(p,\alpha)^{97}\text{Nb}$  reactions have been sparsely characterized and their databases are weak. Experimental work, evaluations and modelling efforts are required to address these particular concerns and inadequacies. The consultants advise that the proton beam energy be kept below 24 MeV to avoid many undesirable impurities. If this requirement is fulfilled, some of the reaction data requested by IACS (e.g., (p,4n), (p,p3n), and to a lesser extent (p,2n)) would be of no concern. Strong communications should be maintained between the Nuclear Data Section and Industrial Applications and Chemistry Section within the IAEA as this type of work evolves.

## 4. ASSESSMENTS AND RECOMMENDATIONS

Various types of radionuclide were considered on the basis of their decay characteristics and possible application in nuclear medicine:

- diagnostic  $\gamma$ -ray emitters,
- positron emitters,
- therapeutic beta, X-ray and  $\gamma$ -ray emitters,
- therapeutic Auger-electron emitters,
- therapeutic alpha emitters.

Cross-section and decay-data studies for various relevant IAEA CRPs and the database of the Decay Data Evaluation Project (DDEP),

[http://www.nucleide.org/DDEP\\_WG/DDEPdata.htm](http://www.nucleide.org/DDEP_WG/DDEPdata.htm)

as well as NuDat retrievals from the ENSDF database

<http://www.nndc.bnl.gov/nudat2/>

were inspected, and used as aids to identify required improvements. The need to provide the user community with uncertainty values in the recommended data was discussed at length. All recommended cross-section data are expected to have uncertainties. However, the feasibility of undertaking such a task should be assessed.



#### 4.1. Diagnostic $\gamma$ -ray Emitters

See Table 1.

Reactor-produced  $^{99}\text{Tc}^{\text{m}}$  is the most commonly used  $\gamma$ -ray emitting radionuclide for diagnostic purposes, and both the cross-section and decay data are well known. The cyclotron-production methods for  $^{67}\text{Ga}$ ,  $^{111}\text{In}$ ,  $^{123}\text{I}$  and  $^{201}\text{Tl}$  have been recently evaluated in detail (IAEA-TECDOC-1211<sup>1</sup>), and their decay data are reasonably well defined. However, both  $^{67}\text{Ga}$  and  $^{111}\text{In}$  possess relatively long half-lives and are rarely used anymore for diagnostic purposes – therefore, they were only considered as therapeutic radionuclides (see Sections 4.3 and 4.4). Discussions focused on recent developments, especially with respect to emerging therapeutic applications of  $^{99}\text{Tc}^{\text{m}}$  and  $^{123}\text{I}$ .

$^{99}\text{Tc}^{\text{m}}$ : new data requirements have arisen as a consequence of recent efforts to produce this radionuclide by means of charged-particle accelerators rather than fission reactors. Various neutron-, proton-, deuteron-, photofission- and photoneutron-induced reactions need to be experimentally studied. Cross-section studies are required, especially with respect to impurities. Auger-electron and other low-energy electron decay data required to assess microdosimetry.

$^{97}\text{Ru}$ : Cross-section measurements and evaluations required for  $^3\text{He}$  and  $^4\text{He}$  beams.

$^{123}\text{I}$ : Auger emissions may become an issue, if a therapeutic application arises in the future.

$^{147}\text{Gd}$ : Cross-section measurements and evaluations required for  $^3\text{He}$  and  $^4\text{He}$  beams.

#### 4.2. Positron Emitters

See Table 2.

The production and decay data of the four most commonly used, short-lived positron emitters in PET studies have been recently evaluated and well quantified ( $^{11}\text{C}$ ,  $^{13}\text{N}$ ,  $^{15}\text{O}$  and  $^{18}\text{F}$  – see IAEA-TECDOC-1211<sup>1</sup>). Nevertheless, further requirements related to proton therapy were considered ( $E_p > 20$  MeV) during the course of the discussions, along with the development and potential adoption of other positron-emitting radionuclides.

$^{57}\text{Ni}$ ,  $^{66}\text{Ga}$ ,  $^{72}\text{As}$ ,  $^{73}\text{Se}$ ,  $^{75}\text{Br}$ ,  $^{76}\text{Br}$ ,  $^{77}\text{Kr}$ ,  $^{81}\text{Rb}$ ,  $^{82}\text{Rb}^{\text{m}}$ ,  $^{83}\text{Sr}$ ,  $^{89}\text{Zr}$ ,  $^{94}\text{Tc}^{\text{m}}$  and  $^{121}\text{I}$ : cross-section and decay-data measurements and evaluations required.

$^{44}\text{Ti}/^{44}\text{Sc}$ ,  $^{52}\text{Fe}/^{52}\text{Mn}^{\text{m}}$ ,  $^{62}\text{Zn}/^{62}\text{Cu}$ ,  $^{72}\text{Se}/^{72}\text{As}$  and  $^{140}\text{Nd}/^{140}\text{Pr}$  generators: cross-section and decay-data measurements and evaluations required.

$^{86}\text{Y}$  and  $^{120}\text{I}^{\text{g}}$ : cross-section evaluations and decay-data measurements and evaluations required.

$^{11}\text{C}$ ,  $^{13}\text{N}$ ,  $^{14,15}\text{O}$ ,  $^{30}\text{P}$  and  $^{38}\text{K}$ : cross-section measurements and evaluations need to be extended to a proton energy of 250 MeV.

$^{95}\text{Ru}$ : cross-section measurements and evaluations required for  $^3\text{He}$  and  $^4\text{He}$  beams.

$^{34}\text{Cl}^{\text{m}}$ ,  $^{43}\text{Sc}$ ,  $^{45}\text{Ti}$ ,  $^{48}\text{V}$ ,  $^{49}\text{Cr}$ ,  $^{51}\text{Mn}$ ,  $^{52}\text{Mn}$ ,  $^{68}\text{Ga}$ ,  $^{90}\text{Nb}$  and  $^{152}\text{Tb}$ : cross-section measurements and evaluations required.

$^{68}\text{Ge}/^{68}\text{Ga}$  and  $^{82}\text{Sr}/^{82}\text{Rb}$  generators: cross-section measurements and evaluations required.

$^{52}\text{Fe}$ ,  $^{55}\text{Co}$ ,  $^{61}\text{Cu}$  and  $^{110}\text{In}^{\text{m}}$ : cross-section evaluations required.

#### 4.3. Therapeutic $\beta^-$ , X-ray and $\gamma$ -ray Emitters

See Table 3.

$\beta^-$ -emitting radionuclides often used to perform internal radiotherapy were not assessed in the current review because they have undergone detailed study in a recent IAEA CRP (see IAEA Technical Reports Series No. 473<sup>2</sup> for the production of  $^{32}\text{P}$ ,  $^{89}\text{Sr}$ ,  $^{90}\text{Y}$ ,  $^{131}\text{I}$ ,  $^{186}\text{Re}$ ,  $^{188}\text{Re}$  and others). Commonly used sources for external radiation therapy (e.g.,  $^{60}\text{Co}$  and  $^{137}\text{Cs}$ ) and brachytherapy (e.g.,  $^{192}\text{Ir}$ ) were also not considered. Focus was placed on  $\beta^-$  emitters with

known discrepant data, and radionuclides believed to possess future therapeutic potential (specifically low-energy  $\beta^-$  and X-ray emitters).

$^{169}\text{Er}$  and  $^{175}\text{Yb}$ : cross-section and decay-data measurements and evaluations required.

$^{47}\text{Sc}$ ,  $^{131}\text{Cs}$ ,  $^{131}\text{Ba}$  and  $^{166}\text{Ho}$ : cross-section measurements and evaluations required.

$^{67}\text{Cu}$ ,  $^{103}\text{Pd}$  and  $^{161}\text{Tb}$ : decay-data measurements and evaluations required.

$^{191}\text{Os}/^{191}\text{Ir}^{\text{m}}$  generator: cross-section measurements and evaluations required.

$^{191}\text{Pt}/^{191}\text{Ir}^{\text{m}}$  generator: cross-section measurements and evaluations required, along with decay-data measurements and evaluation of  $^{191}\text{Pt}$  parent.

#### 4.4. Therapeutic Auger-electron Emitters

See Table 4.

$^{125}\text{I}$  is the most commonly used Auger-electron emitter for internal radiotherapy – both reactor-production and decay data are well characterized.

All of the following radionuclides were identified as potentially suitable for application with respect to microdosimetry at the molecular and cellular levels, and therefore would require much improved Auger-electron decay data:

$^{71}\text{Ge}$ ,  $^{178}\text{Ta}$ ,  $^{193}\text{Pt}^{\text{m}}$ ,  $^{195}\text{Pt}^{\text{m}}$  and  $^{197}\text{Hg}$ : cross-section studies are also required.

$^{67}\text{Ga}$ ,  $^{77}\text{Br}$ ,  $^{99}\text{Tc}^{\text{m}}$ ,  $^{103}\text{Pd}$ ,  $^{111}\text{In}$ ,  $^{123}\text{I}$  and  $^{140}\text{Nd}$ .

#### 4.5. Therapeutic $\alpha$ Emitters

See Table 5.

$^{149}\text{Tb}$ : cross-section measurements and evaluations of spallation and heavy-ion reactions required.

$^{211}\text{At}/^{211}\text{Po}$ : well-established radionuclidic combination for therapeutics –  $^{211}\text{At}$  half-life is rather short at 7.2 h.

$^{225}\text{Ac}/^{213}\text{Bi}$ : cross-section measurements and evaluation of spallation reaction on  $^{232}\text{Th}$  required.

$^{227}\text{Ac}/^{223}\text{Ra}$ : cross-section measurements and evaluation of  $^{232}\text{Th}(p,x)$  reaction to produce  $^{227}\text{Ac}$  required.

$^{230}\text{U}/^{226}\text{Th}$ : cross-section and decay-data measurements and evaluations are required.

#### 4.6. Proton Beam Therapy

Necessary improvements are required with respect to specific cross-section data for proton beam therapy:

- (1) non-elastic cross sections of the light elements (C, N and O) at proton energies up to 250 MeV; and
- (2) activation cross sections of residual nuclei, particularly the positron-emitters  $^{11}\text{C}$ ,  $^{13}\text{N}$ ,  $^{15}\text{O}$ ,  $^{30}\text{P}$  and  $^{38}\text{K}$ .

Owing to the complexity of the fragmentation reactions that occur in a carbon beam, data sets that describe these reactions well would be extremely difficult to prepare. Therefore, the best course of action would appear to be the development of more precise models and validated parameter sets to describe the fragmentation processes and the production of light particles and residues. The derivation of better models and validated parameters would also be the ideal approach for proton beam therapy, in order to undertake more precise Monte-Carlo transport calculations of the effects on dose deposition of variations in morphology or in structure arising from bone or implants.

**Table 1: Diagnostic radionuclides: gamma emitters.**

Radionuclide	Production data	Decay data <sup>†</sup>	CRP <sup>*</sup>	Comments
<sup>99m</sup> Tc	<sup>100</sup> Mo(p,2n): evaluation required. <sup>100</sup> Mo(p,αn) and (p,α): new measurements required before re-evaluation. Impurities: <sup>98</sup> Mo(p,2n) <sup>97m</sup> Tc: measurements required. <sup>100</sup> Mo(p,3n) <sup>98</sup> Tc and (p,4n) <sup>97</sup> Tc: difficult measurements required – theoretical calculations may suffice.	Decay data evaluated in previous CRP (X-ray and Gamma-ray Decay Data Standards). Data required for Auger electrons (E <sub>e</sub> < 25 keV) and other low-energy electrons (E <sub>e</sub> < 1 keV) for microdosimetry (see Table 4).	√	Data requirements to focus on accelerator production – such studies required, especially with regard to impurities. Availability of highly enriched <sup>100</sup> Mo (> 99%) should be investigated.
	<sup>100</sup> Mo(d,3n) <sup>99m</sup> Tc and (d,p2n) <sup>99</sup> Mo – evaluations required.		√	Limited applicability arises from sparse availability of suitable accelerators.
	<sup>100</sup> Mo(n,2n) <sup>99</sup> Mo data: new measurements required for neutron generation by deuterons on carbon targets.		√	(n,2n) reaction product of low specific activity.
	Photofission- and photoneutron-induced reactions to produce <sup>99</sup> Mo.		√	(γ,n) reaction product of low specific activity; (γ,f) reaction more suitable, but has low cross section.
<sup>97</sup> Ru	<sup>3</sup> He and <sup>4</sup> He beams on Mo: measurements and evaluations required.			Limited application.
<sup>123</sup> I	Evaluated in previous CRP (Charged-particle Cross-section Database for Medical Radioisotope Production: Diagnostic Radioisotopes and Monitor Reactions, IAEA-TECDOC-1211, 2001).	Decay data evaluated in previous CRP (X-ray and Gamma-ray Decay Data Standards). Auger emissions may become an issue to be addressed in the future (see Table 4).	√	Several production reactions will be studied in planned CRP (improvements in charged-particle monitor reactions and nuclear data for medical radionuclides, INDC(NDS)-0591, 2011) – observed discrepancies will also be investigated further.
<sup>147</sup> Gd	<sup>4</sup> He beam on Sm, and proton beam on Eu: measurements and evaluations required.	Extremely complex gamma-ray decay scheme.		Special application in MRI + SPECT.
<sup>203</sup> Pb	Production data are known.			Special application in tracer studies.

<sup>†</sup> All decay scheme data should be rigorously assessed to ensure their suitability with respect to decay characteristics, data uncertainties and completeness (see main text).

<sup>\*</sup> Symbol √ denotes relevant studies undertaken as part of either a completed or on-going CRP.

**Table 2: Positron emitters.**

<b>Radionuclide</b>	<b>Production data</b>	<b>Decay data<sup>†</sup></b>	<b>CRP<sup>*</sup></b>	<b>Comments</b>
<sup>11</sup> C, <sup>13</sup> N, <sup>14,15</sup> O, <sup>30</sup> P, <sup>38</sup> K	Measurements and evaluations required of activation cross sections for proton-induced reactions with energies up to 250 MeV.			Cross sections are well defined for proton-induced reactions with E <sub>p</sub> < 20 MeV; however, higher energies up to 250 MeV are of interest for proton therapy.
<sup>34</sup> Cl <sup>m</sup>	Measurements and evaluations required.			Low priority radionuclide.
<sup>43</sup> Sc	Measurements and evaluations required.	Good positron-decay characteristics.		Although difficult to produce, <sup>43</sup> Sc would potentially be very useful.
<sup>45</sup> Ti, <sup>48</sup> V, <sup>49</sup> Cr, <sup>90</sup> Nb	Measurements and evaluations required.			Potentially important for radioimmunotherapy.
<sup>51,52</sup> Mn	Measurements and evaluations required.			Special application in MRI + PET.
<sup>52</sup> Fe, <sup>55</sup> Co, <sup>61</sup> Cu, <sup>110</sup> In <sup>m</sup>	Evaluations required.		√	Several novel applications.
<sup>57</sup> Ni, <sup>72</sup> As, <sup>73</sup> Se, <sup>94</sup> Tc <sup>m</sup>	Measurements and evaluations required.	Measurements and evaluation of positron and X-ray emission probabilities required.	√	Priority 2 decay-data evaluation in planned CRP (improvements in charged-particle monitor reactions and nuclear data for medical radionuclides, INDC(NDS)-0591, 2011).
<sup>64</sup> Cu	Evaluated in previous CRP (Nuclear Data for the Production of Therapeutic Radionuclides, IAEA Technical Reports Series No. 473, 2011).		√	Important positron emitter, especially for radioimmunotherapy.
<sup>66</sup> Ga	Measurements and evaluations required.	Measurements and evaluation of positron and X-ray emission probabilities required.	√	Priority 1 decay-data evaluation in planned CRP (improvements in charged-particle monitor reactions and nuclear data for medical radionuclides, INDC(NDS)-0591, 2011).
<sup>68</sup> Ga	Measurements and evaluations required.		√	Direct production of <sup>68</sup> Ga is attracting attention, as well as the <sup>68</sup> Ge/ <sup>68</sup> Ga generator route, because of increased application.
<sup>75</sup> Br, <sup>77</sup> Kr	Measurements and evaluations required.	Measurements and evaluation of positron and X-ray emission probabilities required.		Limited application.

**Table 2: Positron emitters (cont'd).**

<b>Radionuclide</b>	<b>Production data</b>	<b>Decay data<sup>†</sup></b>	<b>CRP<sup>*</sup></b>	<b>Comments</b>
<sup>76</sup> Br, <sup>89</sup> Zr	Measurements and evaluations required.	Measurements and evaluation of positron and X-ray emission probabilities required.	√	Priority 3 decay-data evaluation in planned CRP (improvements in charged-particle monitor reactions and nuclear data for medical radionuclides, INDC(NDS)-0591, 2011).
<sup>81</sup> Rb, <sup>82</sup> Rb <sup>m</sup> , <sup>83</sup> Sr	Measurements and evaluations required.	Measurements and evaluation of positron and X-ray emission probabilities required.		Limited application.
<sup>86</sup> Y	Evaluations required.	Measurements and evaluation of positron and X-ray emission probabilities required.	√	Important positron emitter for quantification of dosimetry calculations. Priority 1 decay-data evaluation in planned CRP (improvements in charged-particle monitor reactions and nuclear data for medical radionuclides, INDC(NDS)-0591, 2011).
<sup>95</sup> Ru	<sup>3</sup> He and <sup>4</sup> He beams: measurements and evaluations.	Many gamma emissions, together with ~14% positron emission probability.		Limited application.
<sup>120</sup> I <sup>g</sup>	Evaluations required.	Measurements and evaluation of positron and X-ray emission probabilities required.	√	Priority 3 decay-data evaluation in planned CRP (improvements in charged-particle monitor reactions and nuclear data for medical radionuclides, INDC(NDS)-0591, 2011).
<sup>121</sup> I	Measurements and evaluations required.	Many gamma emissions, together with ~11% positron emission.		Easier to produce than <sup>120</sup> I. Borderline - for longer-term consideration.
<sup>124</sup> I	Evaluated in previous CRP (Nuclear Data for the Production of Therapeutic Radionuclides, IAEA Technical Reports Series No. 473, 2011).			Important positron emitter for quantification of dosimetry calculations.
<sup>152</sup> Tb	Measurements and evaluations required.			Potentially useful as lanthanide-based positron emitter.

**Table 2: Positron emitters (cont'd).**

Radionuclide	Production data	Decay data <sup>†</sup>	CRP*	Comments
<b>Generators:</b>				
<sup>44</sup> Ti/ <sup>44</sup> Sc	Measurements and evaluations required.	Evaluation required of parent T <sub>1/2</sub> .		Long-lived parent (T <sub>1/2</sub> of 60 y); difficult to produce.
<sup>52</sup> Fe/ <sup>52</sup> Mn <sup>m</sup>	Measurements and evaluations required.	Measurements and evaluation required.		Special application in MRI + PET.
<sup>62</sup> Zn/ <sup>62</sup> Cu	Measurements and evaluations required.	Measurements and evaluation of positron and X-ray emission probabilities of daughter required.	√	Priority 2 decay-data evaluation in planned CRP (improvements in charged-particle monitor reactions and nuclear data for medical radionuclides, INDC(NDS)-0591, 2011).
<sup>68</sup> Ge/ <sup>68</sup> Ga, <sup>82</sup> Sr/ <sup>82</sup> Rb	Measurements and evaluations required.		√	Well-established systems, but databases inadequate.
<sup>72</sup> Se/ <sup>72</sup> As	Measurements and evaluation required.	Measurements and evaluation of positron and X-ray emission probabilities required.	√	Priority 2 decay-data evaluation in planned CRP (improvements in charged-particle monitor reactions and nuclear data for medical radionuclides, INDC(NDS)-0591, 2011).
<sup>140</sup> Nd/ <sup>140</sup> Pr	Measurements and evaluation required.	Auger-electron and other low-energy electron data required for <sup>140</sup> Nd microdosimetry.		Radiotherapy + PET. Parent <sup>140</sup> Nd(EC) to operate as therapeutic radionuclide, while <sup>140</sup> Pr is a positron emitter ( <i>in-vivo</i> generator).

<sup>†</sup> All decay scheme data should be rigorously assessed to ensure their suitability with respect to decay characteristics, data uncertainties and completeness (see main text).

\* Symbol √ denotes relevant studies undertaken as part of either a completed or on-going CRP.

**Table 3: Therapeutic beta and X-ray/gamma emitters.**

Radionuclide	Production data	Decay data <sup>†</sup>	CRP <sup>*</sup>	Comments
<sup>47</sup> Sc	Measurements and evaluations required.			Low-energy β <sup>-</sup> emitter.
<sup>67</sup> Cu	Evaluated in previous CRP (Nuclear Data for the Production of Therapeutic Radionuclides, IAEA Technical Reports Series No. 473, 2011).	Measurements and evaluation required, particularly with respect to g.s. to g.s. transition.		Important radionuclide – emission of low-energy β <sup>-</sup> particles, and preparation of organometallic complexes.
<sup>103</sup> Pd	Evaluated in previous CRP (Nuclear Data for the Production of Therapeutic Radionuclides, IAEA Technical Reports Series No. 473, 2011).	Data discrepancies – measurements and evaluation required.	√	Priority 1 decay-data evaluation in planned CRP (improvements in charged-particle monitor reactions and nuclear data for medical radionuclides, INDC(NDS)-0591, 2011). Also included as Auger emitter in table below.
<sup>131</sup> Cs	Measurements and evaluations required.		√	X-ray emitter.
<sup>131</sup> Ba	Measurements and evaluations required.	Complex decay scheme requires assessment.	√	X-ray emitter.
<sup>161</sup> Tb	<sup>160</sup> Gd(n,γ) <sup>161</sup> Gd(β <sup>-</sup> ) <sup>161</sup> Tb	Measurements and evaluation required.		Low-energy β <sup>-</sup> emitter.
<sup>166</sup> Ho	Evaluated in previous CRP for reactor production only (Nuclear Data for the Production of Therapeutic Radionuclides, IAEA Technical Reports Series No. 473, 2011). <sup>164</sup> Dy(2n,γ) <sup>166</sup> Dy(β <sup>-</sup> ) <sup>166</sup> Ho: measurements and evaluation required.	<sup>166</sup> Ho decay data evaluated in previous CRP (X-ray and Gamma-ray Decay Data Standards).		High-flux reactor required for double-neutron capture.
<sup>169</sup> Er	Measurements and evaluations required, including spallation beam cross sections.	Measurements and evaluation required.		Low-energy β <sup>-</sup> emitter.
<sup>175</sup> Yb	Measurements and evaluations required for charged-particle reactions.	Measurements and evaluation required.		Low-energy β <sup>-</sup> emitter.
<sup>191</sup> Os/ <sup>191</sup> Ir <sup>m</sup>	Measurements and evaluations required.			Low-energy β <sup>-</sup> emitter for radiotherapy + SPECT. Potential <i>in-vivo</i> generator. Difficult to produce by means of charged-particle reactions.
<sup>191</sup> Pt/ <sup>191</sup> Ir <sup>m</sup>	Measurements and evaluations required.	Measurements and evaluation required of parent decay data.		X-ray emitter. Potential <i>in-vivo</i> generator. Difficult to produce by means of charged-particle reactions.

<sup>†</sup> All decay scheme data should be rigorously assessed to ensure their suitability with respect to decay characteristics, data uncertainties and completeness (see main text).

<sup>\*</sup> Symbol √ denotes relevant studies undertaken as part of either a completed or on-going CRP.

**Table 4: Therapeutic Auger-electron emitters.**

<b>Radionuclide</b>	<b>Production data</b>	<b>Decay data<sup>†</sup></b>	<b>CRP<sup>*</sup></b>	<b>Comments</b>
<sup>67</sup> Ga, <sup>111</sup> In	Evaluated in two previous CRPs (Charged-particle Cross-section Database for Medical Radioisotope Production: Diagnostic Radioisotopes and Monitor Reactions, IAEA-TECDOC-1211, 2001 ( <sup>67</sup> Ga and <sup>111</sup> In), and Nuclear Data for the Production of Therapeutic Radionuclides, IAEA Technical Reports Series No. 473, 2011 ( <sup>67</sup> Ga)).	Auger emissions may become an issue to be addressed in the future.		Previously used in diagnostic studies, both <sup>67</sup> Ga and <sup>111</sup> In are finding increased application in internal radiotherapy.
<sup>71</sup> Ge	Measurements and evaluations required.	Auger emissions may become an issue to be addressed in the future.		Half-life is rather long at 11.4 d.
<sup>77</sup> Br	Several reactions have been investigated – evaluations required.	Auger emissions may become an issue to be addressed in the future.		
<sup>99</sup> Tc <sup>m</sup>	Commonly available – data needs will only arise if produced by charged-particle reactions on accelerators.	Decay data evaluated in previous CRP (X-ray and Gamma-ray Decay Data Standards). Data required for Auger electrons (E <sub>e</sub> < 25 keV) and other low-energy electrons (E <sub>e</sub> < 1 keV) for microdosimetry.		While regularly used for diagnosis, there is increased application in therapeutics.
<sup>103</sup> Pd	Evaluated in previous CRP (Nuclear Data for the Production of Therapeutic Radionuclides, IAEA Technical Reports Series No. 473, 2011).	Measurements and evaluations required.	√	Priority 1 decay-data evaluation in planned CRP (improvements in charged-particle monitor reactions and nuclear data for medical radionuclides, INDC(NDS)-0591, 2011).
<sup>123</sup> I	Evaluated in previous CRP (Charged-particle Cross-section Database for Medical Radioisotope Production: Diagnostic Radioisotopes and Monitor Reactions, IAEA-TECDOC-1211, 2001).	Auger emissions may become an issue to be addressed in the future.		While regularly used for diagnosis, there is increased application in therapeutics.



**Table 4: Therapeutic Auger-electron emitters (cont'd).**

<b>Radionuclide</b>	<b>Production data</b>	<b>Decay data<sup>†</sup></b>	<b>CRP<sup>*</sup></b>	<b>Comments</b>
<sup>140</sup> Nd	Several reactions have been investigated – evaluations required.	Auger emissions may become an issue to be addressed in the future.		Auger and EC decay. <i>In-vivo</i> generator ( <sup>140</sup> Pr) – see Table 2.
<sup>178</sup> Ta	<sup>176</sup> Hf( $\alpha,2n$ ) <sup>178</sup> W(EC) <sup>178</sup> Ta	Auger emissions may become an issue to be addressed in the future.		Auger and EC decay. <i>In-vivo</i> generator ( <sup>178</sup> W).
<sup>193</sup> Pt <sup>m</sup> , <sup>195</sup> Pt <sup>m</sup>	Measurements and evaluations required.	Auger emissions may become an issue to be addressed in the future.		Large number of Auger electrons emitted.
<sup>197</sup> Hg	Measurements and evaluations required.	Auger emissions may become an issue to be addressed in the future. Measurements and evaluation required.		

<sup>†</sup> All decay scheme data should be rigorously assessed to ensure their suitability with respect to decay characteristics, data uncertainties and completeness (see main text).

<sup>\*</sup> Symbol  $\checkmark$  denotes relevant studies undertaken as part of either a completed or on-going CRP.

**Table 5: Therapeutic alpha emitters.**

<b>Radionuclide</b>	<b>Production data</b>	<b>Decay data<sup>†</sup></b>	<b>CRP<sup>*</sup></b>	<b>Comments</b>
<sup>149</sup> Tb	Measurements and evaluations of spallation and heavy-ion beam reactions.			Emission of low-energy alpha particles (< 4 MeV) renders <sup>149</sup> Tb potentially useful for special applications.
<sup>211</sup> At/ <sup>211</sup> Po	Evaluated in previous CRP (Nuclear Data for the Production of Therapeutic Radionuclides, IAEA Technical Reports Series No. 473, 2011).	<sup>211</sup> At and <sup>211</sup> Po decay data evaluated in CRP (Updated Actinide Decay Data Library).		Well-established therapeutic radionuclide.
<sup>225</sup> Ac/ <sup>213</sup> Bi	Lack of cross-section data at higher energies for spallation reaction on <sup>232</sup> Th.	Full decay chain evaluated in CRP (Updated Actinide Decay Data Library).	√	Potentially important therapeutic radionuclide.
<sup>227</sup> Ac/ <sup>223</sup> Ra	Inadequate cross-section database for <sup>232</sup> Th(p,x) production of <sup>227</sup> Ac.	Experimental data and evaluation required for <sup>227</sup> Ac. <sup>223</sup> Ra evaluated in CRP (Updated Actinide Decay Data Library).	√	Impurity in <sup>225</sup> Ac production.
<sup>230</sup> U/ <sup>226</sup> Th	Production reactions will be studied within planned CRP (production and decay-data characteristics of existing and research-based PET radionuclides).	<sup>230</sup> U decay chain requires evaluation - papers containing new measurements will appear in 2012 (following their presentation at ICRM2011 conference).	√	Decay-data evaluations for <sup>230</sup> U decay chain were not included in CRP entitled “Updated Actinide Decay Data Library”.

<sup>†</sup> All decay scheme data should be rigorously assessed to ensure their suitability with respect to decay characteristics, data uncertainties and completeness (see main text).

<sup>\*</sup> Symbol √ denotes relevant studies undertaken as part of either a completed or on-going CRP.

## 5. LONG-TERM PERSPECTIVES

Extremely significant developments are currently taking place in both organ imaging and internal radionuclide therapy. The dynamic and quantitative nature of positron tomography (PET) is being coupled with X-ray tomography (CT) and magnetic resonance imaging (MRI) to provide a highly powerful combination of systems for organ imaging. New possibilities for improved internal radiotherapy are also being assessed and developed in terms of two proposed procedures:

- (1). a combination of PET and therapy involving radioimmuno reactions, and
- (2). Auger-electron and  $\alpha$ -particle therapy at the cellular level.

Future success with these methods of treatment is difficult to gauge, which impacts significantly on the ability to identify long-term nuclear data needs.

With regard to long-term perspectives, participants assessed a relatively large number of radionuclides deemed to be potentially suitable for application in nuclear medicine. Overall, demands are expected to move towards positron emitters and therapeutic radionuclides. The increased adoption of metallic-based positron emitters can be envisaged to occur as a consequence of developments in organometallic-complex chemistry (especially for Ti, Ga and Cu radionuclides), while improvements in microdosimetry techniques may require much improved characterisation of suitable low-energy Auger-electron emitters to support the efficacy of this form of therapeutic treatment.

## 6. CONCLUDING REMARKS

The contents of this report constitute a subjective selection of relevant radionuclides based on our knowledge of emerging developments within nuclear medicine and therapy applications. Participants assessed and reviewed the known cross-section and decay data for a relatively large number of radionuclides deemed as potentially suitable for nuclear medicine. Recommendations focused on the need for cross-section studies over a reasonably wide range of targets and projectiles, along with decay data measurements for specific radionuclides, as described and tabulated in Section 4. The need to provide the user community with uncertainty values in the recommended data was also stressed. All recommended beam-monitor cross sections are expected to have uncertainties. Comprehensive cross-section and decay-data evaluations that include recommended data uncertainties are clearly merited to ensure the necessary quality and consistency of any data assembled in an appropriate database for nuclear medicine applications.

A dissemination system needs to be developed by the IAEA Nuclear Data Section (possibly assisted by external consultants) to handle all recommended decay data for medical applications that arise from any data evaluation and assembly initiative instigated by the IAEA. MIRD could serve as a starting point, but the new system should be extended to include additional data such as low-energy X-rays and Auger electrons. A comprehensive calculational route also needs to be developed to determine the energies and emission probabilities of the low-energy X-rays and Auger electrons to a higher degree of detail and consistency than is available at present. The aim should be to produce a definitive and consistent set of data of all the nuclides of immediate value to the medical profession. Resulting recommended decay schemes should also be used for cross-section and activity measurements.

Radionuclides production in the foreseeable future will require intermediate-energy, high-power accelerators with extended proton energies up to 200 MeV. Small cyclotron systems operating to a maximum of 30 MeV will not suffice for a significant number of the

radionuclides considered in this report. Furthermore, there will be a need for such facilities to generate deuterons,  $^3\text{He}$  and  $^4\text{He}$  particle beams in order to extend the range of production and improve radionuclidic purity. Activation products relevant to proton and heavy-ion radiotherapy also need to be better quantified with respect to their production cross sections (particularly the positron-emitters  $^{11}\text{C}$ ,  $^{13}\text{N}$ ,  $^{15}\text{O}$ ,  $^{30}\text{P}$  and  $^{38}\text{K}$ ).

The discussions and interactions between decay-data and cross-section reaction specialists during the course of this IAEA technical meeting were essential, and assisted greatly in the coverage of key facets of radionuclide usage within diagnostic and therapeutic nuclear medicine. This form of multi-discipline debate should be regularly encouraged by the IAEA to ensure that these important lines of communication are maintained.

Consideration should be given to the organization of a seminar/workshop devoted towards medical radionuclides, organized jointly by the IAC and NDS Sections of the IAEA. The emphasis of this activity should be the use of target chemistry and recommended nuclear data for the production of radionuclides of medical interest. Joint IAEA/ICTP workshops on Nuclear Data for Medical Applications should also be held on a regular basis (every five years).

The IAEA is urged to consider the recommendations contained within this report, as brought together on the basis of the agreed nuclear data requirements for medical radionuclides over the next 5 to 15 years. No attempt was made to prioritize the various nuclear-data requirements identified as important over the specified time scale stretching towards 2025. Rather such efforts should be undertaken at a later date, as IAEA nuclear-data projects in this important area of medical applications are seen to need immediate and substantial development. Plans for such nuclear data projects over this same timescale should take serious note of the requirements and recommendations contained within Sections 3, 4 and 5, above.

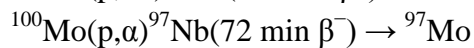
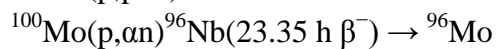
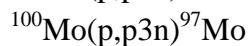
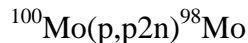
## ANNEX A

### Query and statement prepared by IAEA-IACS staff:

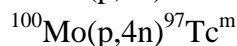
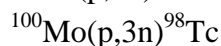
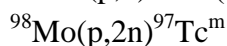
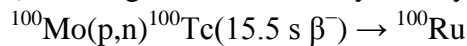
#### Are there still missing data which need to be determined for the large-scale accelerator-based production of $^{99}\text{Tc}^{\text{m}}$ ?

The use of fully enriched  $^{100}\text{Mo}$  targets will never be available, leading to radionuclidic by-products which will always be present and their production rates need careful assessment. Over the proton energy range of 10-25 MeV, several reaction channels still need to be experimentally investigated and compared with theoretical cross-section predictions.

The following reaction channels may have an impact on the isotopic composition of recovered enriched molybdenum and require further assessment:



Also important for further understanding of potential impurities in the final product (including  $^{97}\text{Tc}^{\text{m}}$  which may or may not have dosimetric implications) are:



Moreover, depending on irradiation time and incident beam energy, as well as the energy range (input and output energy), the open reaction channels which contribute to the production of other Tc isotopes will have a direct impact on the  $^{99}\text{Tc}^{\text{m}}$  specific activity.

**See paragraph two of Section 3.3.2 for agreed response of participants attending NDS technical meeting.**





**Technical Meeting on**

**“Intermediate-term Nuclear Data Needs for Medical Applications:  
Cross Sections and Decay Data”**

IAEA Headquarters, Vienna, Austria  
22 – 26 August 2011

Meeting Room A0742

**Preliminary AGENDA**

**Monday, 22 August**

**08:30 - 09:30**     **Registration** (IAEA Registration desk, Gate 1)

**09:30 - 10:15**     **Opening Session**

Welcoming address – Meera Venkatesh (DIR-NAPC)

Introduction – Roberto Capote Noy

Election of Chairman and Rapporteur

Adoption of Agenda

**10:15 - 12:15**     **Presentations by participants**

**12:15 – 12:30**     Administrative matters

*12:30 – 14:00*     *Lunch*

**14:00 – 18:00**     **Presentations by participants (cont'd)**

*Coffee break as needed*

**Tuesday, 23 August**

**09:00 - 12:30**     **Presentations by participants (cont'd)**

*Coffee break as needed*

*12:30 – 14:00*     *Lunch*

**14:00 – 18:00**     **Discussion on proposed future activities – cross-section data**

*Coffee break as needed*

*19:00*                 *Dinner at a Restaurant downtown*

### **Wednesday, 24 August**

**09:00 - 12:30**    **Discussion on proposed future activities – decay data**

*Coffee break as needed*

**12:30 – 14:00**    **Lunch**

**14:00 – 18:00**    **Discussion on proposed future activities - general**

*Coffee break as needed*

### **Thursday, 25 August**

**09:00 - 12:30**    **Drafting of the meeting summary report**

*Coffee break as needed*

**12:30 – 14:00**    **Lunch**

**14:00 – 18:00**    **Drafting of the meeting summary report (cont'd)**

*Coffee break as needed*

### **Friday, 26 August**

**09:00 - 16:00**    **Review of the meeting summary report**

*Coffee and lunch break(s) in between*

**16:00**                **Closing of the meeting**



## APPENDIX 2

### Technical Meeting “Intermediate-term Nuclear Data Needs for Medical Applications: Cross Sections and Decay Data”

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