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INDC International Nuclear Data Committee

Summary Report

Technical Meeting on

Nuclear Data for Medical Applications

IAEA Headquarters Vienna, Austria 10-13 December 2018

Prepared by

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May 2019

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Abstract

A summary is given of a Technical Meeting on "Nuclear Data for Medical Applications" at which participants assessed future medical applications for many radionuclides based upon their existing and potential diagnostic and therapeutic properties. Debate focused upon charged-particle induced reactions and their production cross sections, derivation of optimal yields, minimisation of radionuclidic impurities, and nuclear data needs for proton and heavy-ion radiotherapy, along with outstanding decay data requirements. Technical discussions are included in this report, along with comprehensive listings and detailed recommendations for future work. Required cross-section measurements were identified for a reasonably wide range of targets and projectiles, along with decay data studies for specific radionuclides. Subsequent excitation functions and decay-data evaluations will also be needed to ensure the necessary quality and consistency of the datasets to be assembled in an existing dedicated IAEA-NDS database that merits regular maintenance and support.

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1. INTRODUCTION

Many useful or potentially useful radionuclides have been identified for various diagnostic and therapeutic applications in nuclear medicine. Their most suitable and optimum production routes need to be well defined, along with sound decay scheme data to address dosimetry issues. Nevertheless, deficiencies in such data remain, especially with regard to the optimum production of specific radionuclides, the minimization/elimination of impurities, and the adequate quantification of various decay parameters in specific radionuclides (e.g., half-life, and alpha-particle, electron, positron, gamma-ray and X-ray emission probabilities for comprehensive dose calculations).

Continued developments in medical imaging and therapy utilizing nuclear diagnostic and therapeutic techniques as well as the production of emerging radionuclides call for a further detailed review and expansion of the existing database over an intermediate-term timescale defined as between 5 and 10 years (up to approximately 2029/30). All relevant nuclear data need to be critically reviewed, and new measurements and evaluations recommended if necessary. Therefore, a Technical Meeting on "Nuclear Data for Medical Applications" was held at IAEA Headquarters, Vienna, Austria, from 10 to 13 December 2018, in order to fulfil these requirements. A.J. Koning (Section Head, NDS) welcomed the participants. He emphasized the importance of their role in assessing the nuclear data needs so as to assist greatly in the identification, preparation and characterisation of radionuclides to be used in exploratory and future medical applications. The primary objective of the meeting should be to define potential future nuclear data requirements with the aims of improving preparative routes, radionuclidic purity, and the quantification of various decay characteristics to ensure confidence in considerations of patient dose. IAEA-NDS was represented by R. Capote (Scientific Secretary, Nuclear Data Section). A. Hermanne (Vrije Universiteit Brussel, Brussels, Belgium) was elected Chairman of the meeting, while J.W. Engle (University of Wisconsin, Madison, USA) and A.L. Nichols (University of Surrey, Guildford, UK) served as rapporteurs. The approved Agenda is attached (Appendix 1), as well as a list of participants and their affiliations (Appendix 2).

Atomic and nuclear data are required for both accelerator and reactor production of medical radionuclides, and the current status of such work as organised under the auspices of the IAEA was described in an overview given by R. Capote – presentation available on IAEA-NDS web page <u>www-nds.iaea.org/index-meeting-crp/TM-Med-Apps-2018/</u>. 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-1211¹. 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 handbook² covering reactions used for medically important therapeutic radionuclides.

Following on from the above studies, a consultants' meeting was held on "High-precision Beta-

¹ **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: www-nds.iaea.org/publications/tecdocs/iaea-tecdoc-1211.pdf

² "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.

intensity Measurements and Evaluations for Specific PET Radioisotopes" in September 2008 at IAEA Headquarters, Vienna, Austria³. Other related consultants' meetings have been entitled "Improvements in Charged-particle Monitor Reactions and Nuclear Data for Medical Isotope Production" in June 2011⁴ (re-visit to explore possible improvements to the data of Ref. [1]), and "Intermediate-term Nuclear Data Needs for Medical Applications: Cross sections and Decay Data" in August 2011⁵. Specific recommendations from these three consultants' meetings were brought together in 2011 to formulate and agree the scope, work programme and deliverables of a Coordinated Research Project designed to focus on further improvements to specific charged-particle monitor reactions and nuclear data for the most efficient production and characterisation of medical radionuclides. This programme was defined in terms of five work packages that were mostly concluded in 2017/18 (one publication is still in preparation):

- reference cross sections for charged-particle monitor reactions, *Nucl. Data Sheets* 148 (2018) 338-382; <u>https://doi.org/10.1016/j.nds.2018.02.009</u>;
- recommended nuclear data for medical radioisotope production: diagnostic gamma emitters, *J. Radioanal. Nucl. Chem.* **319**, Issue 2 (2019) 487–531; <u>https://doi.org/10.1007/s10967-018-6142-4;</u>
- recommended nuclear data for medical radioisotope production: diagnostic positron emitters, *J. Radioanal. Nucl. Chem.* **319**, Issue 2 (2019) 533–666; <u>https://doi.org/10.1007/s10967-018-6380-5</u>;
- 4) recommended nuclear data for the production of selected therapeutic radionuclides, *Nucl. Data Sheets* **155** (2019) 56-74; <u>https://doi.org/10.1016/j.nds.2019.01.003</u>;
- 5) selected and recommended atomic and nuclear decay data for medical radionuclides, in preparation (2019).

An extended database containing data from all previous medical-based CRPs is available at:

www-nds.iaea.org/medportal/

and earlier entries have regularly undergone a series of improvements and presentational modifications over the intervening years.

2. PRESENTATIONS

Presentations by the consultants are available on IAEA-NDS web page: www-nds.iaea.org/index-meeting-crp/TM-Med-Apps-2018/

2.1. Monitor Reactions: CRP Input, Outcome, and Lessons Learned from 2014 to 2018, A. Hermanne

Results of the beam-monitoring reactions studied in the most recent IAEA coordinated

³ 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. Available online: <u>www-nds.iaea.org/publications/indc/indc-nds-0535.pdf</u>.

⁴ 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**, September 2011, IAEA, Vienna, Austria. Available online <u>www-nds.iaea.org/publications/indc/indc-nds-0591.pdf</u>

⁵ A.L. Nichols, S.M. Qaim and R. Capote, Technical Meeting on "Intermediate-term Nuclear Data Needs for Medical Applications: Cross Sections and Decay Data", 22-26 August 2011, IAEA report **INDC(NDS)-0596**, September 2011, IAEA, Vienna, Austria. Available online <u>www-nds.iaea.org/publications/indc/indc-nds-0596.pdf</u>

research project from 2012 to 2017 are available in two forms: 2018 article published in *Nuclear Data Sheets* [1.1], and the 2017 update of data tables and figures in the IAEA-NDS medical portal at www-nds.iaea.org/medical/monitor reactions.html.

A primary aim of the CRP was to update and broaden the cross-section database of IAEA-TECDOC-1211 by undertaking a full survey of new literature data, earlier experiments and new dedicated experiments. All of the published datasets were considered for correction with respect to the updated monitor cross sections and nuclear decay characteristics. Each resulting individual set of data was evaluated and either selected or rejected for subsequent fitting by the Padé statistical approach, from which new recommended data with uncertainties were produced for the monitor reactions.

Details on the process of gathering input data were presented, and illustrated with tables and figures on data collection (with the possible need to correct for adjusted nuclear data (abundance of γ lines) and addition or change (min. 7%) of cross-section uncertainties) and selection of data, stressing that the process is not standardized and is essentially based on "subjective choices" by the compiler. All data points of a particular set were selected and only in a few cases were clear outliers or systematic shifted energy points (low energy) proposed for rejection.

After the transfer of all selected data in tabular form with adapted uncertainties to Ignatyuk for Padé fitting, the results were discussed and, if judged appropriate, several re-adopted and/or very recently published data added. Output as presented in the article and database contains all reactions considered in IAEA-TECDOC-1211 (22 reactions) and twelve additional reactions to provide a more uniform base or multiple reactions on each particular target material.

Separate Sections in the article are devoted to a particular particle beam, and starts with a table of nuclear data for the activation products which indicates the different references and new evaluations undertaken during the course of the CRP. The text for each individual reaction includes the reason for consideration, the list of all references and rejected references (with reasons for rejection), and hence the datasets used for each fitting, and the parameters of every fit. Evolving changes of the uncertainties as a function of beam energy and a short overview of the contributing reactions are also given. The database was updated in the same format as that used in the on-line version of IAEA-TECDOC-1211 (2007 update), including a tabulated version of the recommended cross sections in 1-MeV energy steps.

The main lessons learned personally are that team work is required for all of the steps (input, data selection, analysis of fits, and publication), and that respect of commitments by participants in such a research programme is essential. "New" datasets are not necessarily better than earlier published values, and that the selection process is not unique, nor a standard procedure. Finally, we have shown based on our new publications that the newly recommended values can adequately be used for beam monitoring, and that differences between old and new values are within the estimated uncertainties.

[1.1] Reference Cross Sections for Charged-particle Monitor Reactions,
A. Hermanne, A.V. Ignatyuk, R. Capote, B.V. Carlson, J.W. Engle,
M.A. Kellett, T. Kibédi, G. Kim, F.G. Kondev, M. Hussain, O. Lebeda,
A. Luca, Y. Nagai, H. Naik, A.L. Nichols, F.M. Nortier, S.V. Suryanarayana,
S.Takács, F.T. Tárkányi and M.Verpelli, *Nucl. Data Sheets* 148 (2018) 338–382.

2.2. Atomic and Nuclear Decay Data for Medical Applications, A.L. Nichols

Requirements for atomic and nuclear decay data were last addressed within the IAEA Nuclear Data Section just over seven years ago in August 2011⁵. Both cross-section and decay data needs were identified on the basis of existing procedures, developing preclinical trials and longer-term potential. Under such ever-changing circumstances, the resulting request list that evolved from this exercise was not expected to be exhaustive and will change with the evolving needs of the medical community. A re-visit of such needs in late 2018 was most welcome after 7.3 years.

Experimental determinations of cross sections and excitation functions of chargedparticle reactions for radionuclide production rely mostly on the well-defined bombardment of a suitable stacked-foil target with specific particle beams to induce activation, followed by identification and quantification of the induced activity mainly by means of γ -ray spectroscopy. The transformation of count rates obtained directly from γ -ray spectroscopy to the activity produced at a reference time relies on a sound knowledge of the emission probabilities (intensities) of appropriate γ lines and on the half-life of the radionuclide in order to correct for decay during the irradiation, cooling and measurement times. However, the decay data used by the authors of such experimental studies are frequently not always the most up to date and can vary due to the fact that recognised sources of such data may well recommend different values.

Unfortunately, the limited availability of CRP effort resulted in somewhat extended delivery times for decay-data evaluations that could only operate in parallel with the equivalent cross-section studies, which made the direct application of relevant X- and γ -ray data impractical. This problem needs to be overcome by undertaking IAEA-sponsored decay-data studies before the equivalent cross-section measurements. Furthermore, any additional, strongly supportive decay-data measurements are essential before any attempted equivalent in-depth evaluations. Nevertheless, despite these difficulties, all such decay-data data derived during the course of the CRP will be included in the IAEA Medical Portal.

Relevant decay-data assessments and evaluations undertaken during the course of the 2012 to 2017 coordinated research project to various degrees of complexity or adopted from other recent IAEA-CRP databases were as follows:

- beam monitors: ⁶¹Cu, ⁶³Zn and ⁶⁶Ga;
- β^+ emitters: ⁵²Mn, ⁶¹Cu, ⁶⁶Ga, ⁷³Se, ⁷⁶Br, ⁸⁹Zr, ^{94m}Tc and ¹²⁰I;
- β^+ emitters, generator systems: ⁴⁴Ti/⁴⁴Sc (⁴⁴Ti half-life only) and ⁵²Fe/^{52m}Mn;
- diagnostic β^+ and therapeutic β^- emitter (dual purpose): ⁶⁴Cu;
- therapeutic β^- and X-ray/ γ emitter: ⁶⁷Cu (also decay-data measurements);
- the rapeutic α emitters:

```
<sup>227</sup>Th/<sup>223</sup>Ra decay chain – <sup>223</sup>Ra, <sup>219</sup>Rn, <sup>215</sup>Po and <sup>211</sup>Bi;
<sup>229</sup>Th decay chain – <sup>225</sup>Ra, <sup>225</sup>Ac and <sup>221</sup>Fr;
```

 230 U decay chain $-^{230}$ U, 226 Th, 218 Rn, 214 Po, 210 Pb, 210 Bi and 210 Po;

• existing or potential therapeutic Auger-electron emitters: pure ^{99m}Tc, ¹⁰³Pd (^{103m}Rh), ¹¹¹In, ¹²⁵I, ¹³¹Cs and high-spin ¹⁷⁸Ta.

Two IAEA-CRP publications of note with respect to contributing to the above embryonic database are "Update of X-ray and γ -ray Standards" (1998-2005)

www-nds.iaea.org/xgamma_standards/

and "Updated Library of Recommended Actinide Decay Data, 2011" (2005-2011) www-nds.iaea.org/publications/tecdocs/sti-pub-1618.pdf An equivalent further set of radionuclides remains to be assessed from the August 2011 studies to determine whether their decay-data requirements are still valid (or may need to be removed from the original request list), while some inadequacies still remain to be addressed as determined from the earlier work listed above:

- diagnostic γ emitters: ¹²³I (Auger electrons?) and ¹⁴⁷Gd (MRI + SPECT);
- β^+ emitters: ⁵⁷Ni, ⁷²As, ^{75,76}Br, ⁷⁷Kr, ^{81,82m}Rb, ⁸³Sr, ⁹⁵Ru and ¹²¹I;
- β^+ emitters, generator systems: ${}^{62}Zn/{}^{62}Cu$, ${}^{72}Se/{}^{72}As$ and ${}^{140}Nd/{}^{140}Pr$;
- therapeutic β⁻ and/or X-ray/γ emitter: ¹³¹Ba, ¹⁶¹Tb, ¹⁶⁹Er, ¹⁷⁵Yb and ¹⁹¹Pt (parent of ^{191m}Ir);
- therapeutic α emitters: ²²⁷Ac/²²⁷Th;
- therapeutic Auger-electron emitters: ⁶⁷Ga, ⁷¹Ge, ⁷⁷Br, ¹²³I, ¹⁴⁰Nd, ^{193m,195m}Pt and ^{197m,197}Hg.

This above list in particular needs to be re-assessed during the course of this technical meeting on the basis of either re-fortification or individual outright rejection. Furthermore, the production and minimization of impurity radionuclides needs to be given much closer attention than previously.

Consideration should also be given to other known publications, particularly the following: "Long-term Needs for Nuclear Data Development", ed: A. Plompen, IAEA report INDC(NDS)-0601, January 2012 – effectively adds to and lends support to the above; "Nuclear Physics for Medicine", ed: F. Azaiez, *et al.*, NuPECC report, ISBN 978-2-36873-008-9, 2014 – extensive summary review rather than a source of nuclear data needs; "Nuclear Data Needs and Capabilities for Applications", USA whitepaper, ed. L.A. Bernstein, *et al.*, LLNL report LLNL-CONF-676585, 2015 – policy document for US Nuclear Data Program, with lists of nuclear data needs in Appendix B; and recent review publications by S.M Qaim, *et al.*

- J. Radioanal. Nucl. Chem. **305** (2015) 233-245 nuclear data for commonly used diagnostic and therapeutic radionuclides;
- Nucl. Med. Biol. 44 (2017) 31-49 present status and future needs;
- ND2016, EPJ Web of Conferences 146 (2017) 08001 present status and future needs;
- J. Radioanal. Nucl. Chem. **318** (2018) 1493-1509 theranostic pairs.

A healthy number of precise decay data measurements have been performed over recent years, e.g., 67 Cu, 82 Rb, 89 Zr, 124 I, 177 Lu, 186 Re, 230 U and progeny, 227 Th, 223 Ra and progeny, and 229 Th, 225 Ac and progeny. Certainly, more laboratory work would be welcome to clarify some of the more complex decay schemes, particularly but not exclusively with respect to β^+ emitters. Such measurements as those performed by the ANL nuclear data group (67 Cu) and the NNDC-BLIP decay data collaboration (82 Rb and equivalent ongoing studies) are to be warmly commended and encouraged. And finally, the advent of molecular dosimetry underlines the need for better defined low-energy Auger-electron emitters for therapeutic applications – potential candidates have been identified, and the challenge of quantifying their most relevant atomic decay characteristics needs to be addressed.

2.3. Nuclear Data Needs for Medical Applications Using Deuteron Breakup Neutrons, Y. Nagai

Cross-section results were presented for the production of ⁹⁹Mo and ⁶⁷Cu by means of the ¹⁰⁰Mo(n,2n)⁹⁹Mo and ⁶⁸Zn(n,np+d)⁶⁷Cu reactions, with neutrons generated from the deuteron breakup reaction on carbon. Two groups have measured the neutron flux of the

deuteron breakup reaction on carbon at a deuteron energy of 40 MeV, and results from both datasets show a continuous neutron-energy spectrum from thermal energy to approximately 40 MeV with a most probable energy of 14 MeV. Neutrons are emitted at forward angles with respect to the deuteron beam direction, and therefore the breakup neutrons can be effectively used to produce medical radionuclides.

The cross section of the 100 Mo(n,2n) 99 Mo reaction has been previously measured to be as large as approximately 1 to 1.5 barn at neutron energies between 10 and 20 MeV. A 100 g sample of MoO₃ was irradiated with neutrons provided by the C(d,n) reaction at the AVF cyclotron at the Cyclotron and Radioisotope Center, Tohoku University, in order to determine the yield of 99 Mo [3.1, 3.2]. The measured yield of 99 Mo agrees well with the numerical result estimated by means of the latest neutron data, which are a factor of two larger than other existing data. Such a result establishes an important finding with respect to the domestic production of 99 Mo: approximately 50% of the existing demand for 99 Mo in Japan could be produced by means of a single suitable accelerator irradiating a 100 g sample of 100 MoO₃ with neutrons generated by 40 MeV, 2mA deuteron breakup reaction on carbon.

Owing to the low availability of ⁶⁷Cu, there have been few medical studies with this particular radionuclide. We have produced ⁶⁷Cu by the combined ⁶⁸Zn(n,np+d) reactions at the Takasaki Ion Accelerators for Advanced Radiation Application, NIQST [3.3]. High-quality ⁶⁷Cu was satisfactorily generated with a minimum level of ⁶⁴Cu impurity. A high uptake in the tumor of ⁶⁷Cu was found, although the specific activity was low owing to the available neutron intensity. The observed accumulation of ⁶⁷Cu in the tumor suggests that ⁶⁷CuCl₂ can be a potential radionuclide agent for cancer radiotherapy. Further therapeutic studies on small animals at an increased dose of ⁶⁷Cu as produced jointly by the ⁶⁸Zn(n,np+d)⁶⁷Cu reactions could be suitably performed by means of existing intense neutron facilities.

- [3.1] F. Minato, K. Tsukada, N. Sato, S. Watanabe, H. Saeki, M. Kawabata, S. Hashimoto and Y. Nagai: J. Phys. Soc. Jpn. 86, (2017) 093201.
- [3.2] K. Tsukada, Y. Nagai, K. Hashimoto, M. Kawabata, F. Minato, H. Saeki, S. Motoishi and M. Itoh: J. Phys. Soc. Jpn. 87 (2018) 043201.
- [3.3] Y. Sugo, K. Hashimoto, M. Kawabata, H. Saeki, S. Sato, K. Tsukada and Y. Nagai: J. Phys. Soc. Jpn. 86 (2017) 023201.

2.4. Addressing Nuclear Data Needs of the US DOE Isotope Program, F.M. Nortier

The US DOE Isotope Program network consists of fifteen participating institutions across the United States of America. Irradiation facilities for production of medical and nonmedical radionuclides follow several approaches. Three participating reactors utilize neutron-capture reactions employing thermal and epi-thermal neutrons (HFIR at Oak Ridge National Laboratory, MURR at the University of Missouri, and ATR at Idaho National Laboratory), while three high-energy accelerator facilities utilize a wide variety of nuclear reactions induced by high-energy proton beams and high-energy secondary neutron fluxes (IPF at Los Alamos National Laboratory, BLIP at Brookhaven National Laboratory, and LEAF at Argonne National Laboratory). Furthermore, six universitybased cyclotron facilities participate, utilizing lower energy charged-particle beams. Under such circumstances, extensive nuclear data are needed by the Program to inform and support radionuclide production of a wide variety of radionuclides via the various production modalities.

A list of Nuclear Data Needs of the Program was compiled in 2016 and updated in 2017 and 2018. More than 200 nuclear reactions are presently identified, for which nuclear cross-section data are needed to inform on the production of more than 60 primary radionuclides, about half of which are used in medical applications. The largest data gaps appear to be in photonuclear cross sections and in the high-energy regions of protonand neutron-induced reactions. US-based efforts to start addressing these needs were initiated in 2010 with the establishment of a production cross-section measurement capability at LANL for protons up to 800 MeV. Since then measurement campaigns for proton-induced excitation functions in Th, Fe, Cu and Nb targets have been conducted. Other campaigns are ongoing (^{nat,121}Sb+p and La+p) and a new campaign is planned (As+p). The latter measurement campaign is part of a new initiative to establish additional measurement capabilities at Lawrence Berkeley National Laboratory (LBNL) and BNL in order to better cover the proton energy range up to 200 MeV. Other initiatives include the development of neutron-induced excitation function measurement capabilities at LBNL using quasi-monoenergetic neutron beams up to 60 MeV and a photonuclear cross-section measurement capability by ANL using near monoenergetic photons up to 50 MeV at the High Intensity Gamma-ray Source (HIyS) of Duke University.

2.5. Novel Medical Radionuclides, QMN Neutrons, and Thick Target Yields, J.W. Engle

Cyclotron Research Group staff at the University of Wisconsin recognize the need for further work to measure and evaluate nuclear reaction and decay data. These requirements occur primarily in the context of current research efforts to produce novel diagnostic and therapeutic radionuclides for medical application by means of low-energy cyclotrons and high-energy national facilities. A primary aim of such work at the University of Wisconsin is to characterize spallation neutron-induced reactions, and determine their potential for radionuclide production, in collaboration with Los Alamos National Laboratory (LANL), Lawrence Berkeley National Laboratory (LBNL), and the iThemba Laboratory in South Africa. These studies have and will continue to focus on (dis)agreements between measured formation excitation functions and experimental thick target yields.

A full description was given of the efforts by the Wisconsin Cyclotron Research Group to provide ⁶⁴Cu, ⁸⁹Zr, ^{52g}Mn, ⁸⁶Y, ^{76/77}Br and many other radionuclides to colleagues and collaborators in the USA via overnight shipments. Despite their increasing clinical relevance, the production routes for some of these radionuclides as applied directly to patients have not yet been evaluated (e.g., ⁶⁴Cu). Similarly, while fast neutrons are of increasing interest for radionuclide production, necessary energy differential measurements of the formation reactions can only be made by the application of quasimonoenergetic neutron beams that are usually produced by the ${}^{7}Li(p,n){}^{7}Be$ nuclear reaction at only a few facilities worldwide. New lithium targets have recently been produced at LANL, installed at LBNL, and irradiated with activation foil targets to characterize the products of neutron-induced reactions. Additionally, beam time has been awarded in 2019 to experimenters from Wisconsin at iThemba Laboratory for further such experiments. Finally, the routine use of production-scale targets at Wisconsin motivates the frequent comparison of experimentally measured yields with calculated yields from measured and evaluated excitation functions, often illuminating discrepancies that have historically proven challenging to explain, and merit further investigation.

2.6. Development of Novel Radionuclides for Medical Applications, S.M. Qaim

Several relevant review articles that focus on radionuclide production have been written in recent years by the author of this presentation. Note was taken that the production technology of radionuclides commonly used in patient care studies for diagnosis and therapeutics is generally well established. However, there is a constant need to develop novel radionuclides, either longer lived non-standard β^+ emitters for investigating slow biological processes, or low-range highly-ionising corpuscular radiation emitters for internal targeted therapy. The four basic pillars of such development work consist of nuclear data, high-current targetry, chemical processing and quality assurance of the product. A radionuclide will find practical application in medicine only if all the four pillars have been successfully accomplished – the emphasis in this talk had obviously been placed on nuclear data. Recent IAEA efforts to provide evaluated cross-section data for the production of novel radionuclides were also praised. However, in a few cases, the existing cross-section database is known to be weak such that evaluation is not meaningful, while any empirical selection of inadequate experimental data remains very controversial.

Non-standard β^+ emitters are often produced by means of low-energy reactions at medical cyclotrons, and the (p,n) reaction on a highly-enriched target isotope is commonly used. A new strategy to produce non-standard β^+ emitters at medical cyclotrons involves the development of a solution target, although this approach requires a strong database close to the threshold of the reaction which is often missing. Therefore, new careful measurements are needed near to the threshold. On the other hand, several β^+ emitters can only be produced via intermediate energy reactions, and therefore detailed cross-section measurements in the proton energy range of up to about 120 MeV are required.

Efforts have intensified over recent years to produce novel therapeutic radionuclides, emitting low-energy β^- particles (e.g., 47 Sc, 67 Cu and 186 Re), α -particles (e.g., 149 Tb and 225 Ac) and low-energy electrons (e.g., 117m Sn, 193m Pt and 195m Pt). Production methods for some of those radionuclides were reviewed, and emphasis was placed on the need to continue research related to the cross-section data of nuclear reactions induced by charged particles, fast neutrons and high-energy photons.

Some new directions in radionuclide applications were discussed, including multimode imaging (e.g., combination of PET and MRI), theranostic approach (i.e., combination of PET and targeted therapy) and radioactive nanoparticles. Relevant radionuclides were considered. Finally, a brief description was given of emerging facilities for radionuclide development work at FZ Jülich.

2.7. Cross-section Measurements of Proton-induced Reactions for the Production of ⁶⁷Cu and ⁴⁷Sc, G. Pupillo and L. Mou

The LARAMED (<u>LA</u>boratory of <u>RA</u>dionuclides for <u>MED</u>icine) project at INFN-LNL is focused upon proton-induced cross-section measurements, following the installation of a 70 MeV cyclotron in 2015 along with appropriate on-going developments in the laboratory facilities and provision of dedicated beam lines. Among radionuclides of major interest in this work programme, we are considering the optimum production of 67 Cu and 47 Sc as long-lived theranostic radionuclides that can be paired with shorter-lived β^+ emitters such as 64 Cu and 43 Sc/ 44 Sc, respectively.

Collaborative studies in conjunction with the Arronax facility have involved the use of the stacked-foils technique and adoption of $^{nat}Al(p,x)^{24}Na$ monitor reaction data provided by IAEA-NDS (up-dated in August 2017). Our resulting cross-section measurements have

included the following reactions:

- ⁶⁸Zn(p,x)⁶⁷Cu,⁶⁷Ga,⁶⁶Ga cross sections in the proton-beam energy range of 35-70 MeV – results were published in January 2018;
- ⁷⁰Zn(p,x)⁶⁷Cu,⁶⁴Cu,⁶⁷Ga,⁶⁶Ga cross sections in the proton-beam energy range of 45-70 MeV – results under review;
- $^{nat}V(p,x)^{47}Sc$, ^{46}Sc , ^{44}Sc , ^{44m}Sc , ^{48}Sc , ^{43}Sc , ^{48}V , ^{43}K , ^{48}Cr , ^{49}Cr , ^{51}Cr cross sections in the proton-beam energy range of 45-70 MeV (preliminary results) we are also going to measure the production of these radionuclides from enriched metal targets of ^{48}Ti , ^{49}Ti and ^{50}Ti as irradiated similarly by proton beams (E_{max} of 70 MeV). As stated earlier, $^{nat}Al(p,x)^{24}Na$ and $^{nat}Ni(p,x)^{57}Ni$ monitor reaction data as provided by the IAEA-NDS (and up-dated in August 2017) will be used to monitor the beam flux in proton-beam energies of interest ($E_p < 70$ MeV).

Collaborative studies with nuclear modelling experts have also been conducted (L. Canton and A. Fontana from INFN, Padova and Pavia) to compare our experimental data with theoretical predictions obtained from three different codes – TALYS, EMPIRE and Fluka (see Subsection 2.13, below).

On the basis of our studies and the goals of this IAEA-NDS technical meeting, we propose that the following immediate actions and longer-term activities be undertaken:

- ensure that uncertainties in the data for the recommended monitor reactions are made available as soon as possible, e.g., ^{nat}Al(p,x)²²Na, ²⁴Na and ^{nat}Ni(p,x)⁵⁷Ni;
- ensure that uncertainties in the recommended cross-section reactions are made available as soon as possible, e.g., ⁶⁸Zn(p,2p)⁶⁷Cu, ⁷⁰Zn(p,x)⁶⁷Cu;
- undertake an evaluation of the $^{nat}V(p,x)^{47}Sc$ reaction (many data are available);
- undertake a re-evaluation of the recommended cross-section data for the 68 Zn(p,2n) 67 Ga reaction at proton-beam energies $E_p > 35$ MeV.

2.8. Nuclear Data Needs Related to Hadron Therapy, A. Ferrari

Production of β^+ emitters by proton, ⁴He and ¹²C beams is a critical ingredient for hadrontherapy monitoring by means of PET techniques. PET monitoring during patient treatment is required to minimize physiological washout, and will emphasize the contribution of short lived β^+ emitters for which data are scarce.

- Some new preliminary data about β^+ emitter production by p and ¹²C beams on carbon (measured ¹⁰C, ¹¹C) and oxygen (measured ¹⁵O) targets were discussed.
- Need for more data were underlined, particularly for ¹²C beam fragmentation on carbon and oxygen target.
- Lack of β^+ emitter production data for ⁴He beam on C, N and O was also underlined as a major issue.

Detailed angle/energy cross sections for fragment production for ¹²C and ⁴He are in great demand for clinical applications:

- these data are also required for the proposed monitoring of therapy by detecting secondary charged particles escaping from the patient;
- similar data will be required for ion radiography/CT, extended to projectile energies large enough to go through a patient.

⁴He beams are going to be in clinical use in the near future:

- attenuation curves and/or total reaction cross sections are a critical ingredient, their error reflecting directly in the dose distributions, particularly in the Bragg peak region and in the fragmentation tail;
- some new data were presented for ⁴He on C and O, for which errors are still large and only the fragmentation channels have been measured;
- fragmentation of ⁴He into ³H/³He requires clarification in view of past contradictory data.

More distant in the future, there is the possibility of adopting ¹⁶O beams and, even more speculative, ¹¹C or ¹⁵O beams which would directly provide the monitoring signal. Therefore, detailed data about the fragmentation cross section will be needed.

The FOOT Collaboration has setup an experiment which is capable of taking accurate double-differential data on a large fragment and angular range in both inverse (for target fragmentation measurements) and direct kinematics (for projectile fragmentation). The experiment plans are to use a variety of beams (available at HIT and GSI) and targets of relevance for hadron-therapy applications. These kinds of data are badly needed for already existing clinical beams, p and ¹²C, and for ⁴He, and the experiment could also provide some of the β^+ production data mentioned above.

The GSI group involved in the ESA Rossini project underlined that data taken at the Trento therapy centre with protons show significant discrepancies between measured neutron ambient dose equivalent values and code predictions. They suggest further work in measuring neutron double-differential yields produced by protons on light targets.

The CERN-Medicis facility (<u>Medical Isotopes Collected from ISOLDE</u>) was also briefly introduced, which is designed to produce unconventional radionuclides for medical research.

2.9. Argentine Project to Develop the Production of ²²⁵Ac, ²¹³Bi and Suitable Radiopharmaceuticals for Therapy: Nuclear Data Needs, S. Siri

The Alpha Project of the National Atomic Energy Commission of Argentina (CNEA) has been formulated and approved to develop a production route for ²²⁵Ac and ²¹³Bi from ²²⁶Ra targets by means of a medium-low energy cyclotron. There are two main phases in the work programme: (i) local development of the technology for optimum production, and (ii) implementation of effective commercial production. Considering the current and foreseeable future demands based on the latest clinical trials and their results, the primary goal of the Alpha Project is to secure sufficient supply of ²²⁵Ac and ²¹³Bi for our country and regions beyond in order to ensure on-going and future treatment of cancer patients and support R&D for new applications.

Production of ²²⁵Ac from ²²⁶Ra is a very challenging technology that will require experience and expertise in several fields. Our country started the construction of a new dedicated nuclear facility in 2016 in accordance with the standards of the nuclear industry, so as to fulfil the goals of the Alpha Project. Still under construction, this installation is expected to become fully operative in 2020/2021, while the first radiochemical laboratories will become operational within the next few months. When fully in production, the facility has been designed to deal with radioactive ²²⁶Ra targets and all resulting gaseous ²²²Rn emissions. Cold laboratories, and radiochemical facilities with appropriate hoods and glove boxes will be available for the various operational procedures. Several hot cells will also be used to handle and process the irradiated materials and their products: one multipurpose hot cell and five hot cells dedicated to the different steps of the process to obtain the desired quality of ²²⁵Ac (two dedicated to the

safe handling of 226 Ra, and also one GMP hot cell (<u>G</u>ood <u>M</u>anufacturing <u>P</u>ractice)). This installation will also possess a pilot plant for the production of small batches of radiopharmaceuticals, initially under local GMP conditions but capable of being upgraded to more demanding international GMP regulations.

²²⁶Ra targets will be irradiated in the Cyclotron Corporation CP42 (25-42 MeV) cyclotron located at the Ezeiza Atomic Centre of CNEA within the fully automated irradiation station for a solid target with the target holder designed to work with sealed ²²⁶Ra targets. Several energy ranges around the maximum of the cross section will be studied in order to explore yields and radionuclidic impurities as a function of the energy range, with special attention paid to ²²⁶Ac, which is a potential byproduct. The R&D phase will involve a holder that has been designed for sealed ²²⁶Ra targets to operate at a low beam power density with 4π water cooling and a maximum of 60 mg of ²²⁶RaCl₂. A dummy system will be tested in the next few months. Accurate quality control will be applied by means of α and γ spectrometry and ICP-MS to determine the radionuclidic purity arising from the production of ^{226}Ac along with ^{225}Ac . Theoretical considerations based on the energy range of the proton beam in the activation process indicate that a cooling time of at least 240 h (one $T_{1/2}$ of ²²⁵Ac) before processing the target in order to decrease the ²²⁶Ac activity to approximately 0.3% of its initial value, giving a ratio ²²⁶Ac/²²⁵Ac of approximately 0.005. This cooling time also accounts for the decay of ²²⁶Ac daughters with the exception of ²²⁶Ra which will be separated during the ²²⁵Ac purification process. This cooling time is based on theoretical data for the cross section of the ${}^{226}Ra(p,n){}^{226}Ac$ reaction at several energies because there are no available experimental data. Theoretical calculations of the yield for the 226 Ra(p,2n) 225 Ac reaction indicate that for the R&D target designed to run at a lower density current (< 100 μ A/cm²) and activated in the range 24-10 MeV, 58 mg ²²⁶RaCl₂ irradiated 24 h at 50 µA will produce 335 mCi EOB per batch, and 167.5 mCi after a cooling time of 240 h. A production target with a greater surface area capable of functioning at 200 µA will require a larger amount of RaCl₂ (230 mg over 2 cm²) with an activation process of 24 h on the same energy range of 24-10 MeV to produce 1.34 Ci of ²²⁵Ac EOB per batch, and 670 mCi after 240 h cooling time. When considering prostate cancer therapy applied at 0.5 mCi/patient with respect to the production site, this amount represents 1340 single patient treatments, wherein losses during the purification process have been ignored. Therefore, a cyclotron run of this reaction once per week, 4 weeks per month, 11 months per year would produce 29.5 Ci of ²²⁵Ac per year (240 h cooling time after irradiation) with the potential to administer to almost 59,000 patient per year.

This amount of ²²⁵Ac needs to be of high radionuclide quality, and would be produced from only one cyclotron running such a proton-beam irradiation once per week. Depending on the evolution of the treatments by alpha-targeted therapy, such a project has the potential to produce enough ²²⁵Ac to achieve our planned goal to secure sufficient supply in support of wide use in patient treatment along with the provision to satisfy R&D needs for emerging new applications as well.

2.10. Measurements of Production Yields of Positron Emitters Relevant for Beam Range Verification in Proton Therapy, C. Guerrero, J.M. Quesada and T. Rodríguez-González

An intensive research programme in proton therapy has focused on *in vivo* range verification. *In-vivo* PET range verification relies on the comparison of measured and estimated activities from β^+ emitters induced by proton-beam activation of the most

abundant elements in the human body, immediately after (long-lived β^+ emitters ¹¹C, ¹³N and ¹⁵O) or during (short-lived β^+ emitters ¹⁰C, ¹²N, ²⁹P and ^{38m}K) the irradiation. The accuracies of the estimated distributions are basically those of the underlying cross sections. However, a revision of the experimental data available in EXFOR has shown that the reaction cross sections of interest have not been measured over the full energy range required (up to 230 MeV), and that there are sizeable discrepancies between the different datasets. Indeed, several studies confirm the need for more accurate measurements, especially in the case of the short-lived β^+ emitters for which there are no data above a proton-beam energy of 55 MeV.

Under such circumstances, an ambitious experimental programme has been launched to expand and improve our knowledge of the production yields of the reactions resulting in the short- and longer-lived β^+ emitters mentioned above. A new method has been developed for the longer-lived isotopes which combines a multi-foil technique with the measurement of the induced activity by means of a PET scanner. This procedure has been successfully tested below 18 MeV at CNA, Spain, and during 2019 will be used at a clinical beam to obtain data up to 230 MeV. However, this method is not suitable for measuring short-lived positron emitters because rapid decay occurs before one can place the films in the PET scanner. Therefore, a set-up combining a stack of targets sandwiched between converter/degrader foils with an array of LaBr₃ detectors is in the process of being designed and developed. Tests below 18 MeV are underway at CNA, while studies at higher energies will be performed at KVI-CART. Both the short- and longer-lived positron emitters require crucial IAEA-CRP evaluated cross-section data for the monitor reactions and the ^{nat}Cu(p,x)⁶³Zn reaction in particular, which is used for normalization purposes.

2.11. New Measurements of the Absolute Auger-electron Yields of Medical Radionuclides, and Recent Developments of the BrIccEmis Code, *T. Kibédi*

Co-workers: M. Vos¹, B. Tee¹, B.Q. Lee^{1,2}, M. Alotiby¹ and I. Greguric³

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Unstable atomic nuclei release excess energy through various radioactive decay processes by emitting radiation in the form of either particles (neutrons, alpha and beta particles) or electromagnetic radiation (photons). Most applications of medical radionuclides make use of the fact that the interaction of the radiations passing through a material depend on their type (photons, neutral or charged particles) and energy. Alpha and beta particles are two well-known ionizing radiations, while a third type of radiation involves the emission of atomic electrons following nuclear decay. Due to their extremely short range, Auger electrons offer subcellular targeting of cancer cells.

BrIccEmis is a Monte-Carlo code that has been recently developed to describe the full energy spectrum of Auger electrons and X-rays following radioactive decay processes [11.1]. The model uses the Evaluated Nuclear Structure Data File (ENSDF) [11.2] to evaluate primary atomic vacancies following electron capture and internal conversion. Atomic transition rates are taken from the Evaluated Atomic Data Library (EADL) [11.3] and atomic binding energies from relativistic Dirac-Fock calculations [11.4]. The model has been successfully used for a range of radionuclides, including several medical radioisotopes studied in a recent IAEA coordinated research project [11.5].

High-resolution electron measurements have been undertaken at the Australian National University to study the low-energy electron spectrum of ^{125}I (see Fig. 1) and subsequently to benchmark the model (^{125}I is also commonly used in nuclear medicine). A thin monolayer source was prepared to measure the electron spectrum in the 0 to 35 keV energy range with an energy resolution of 5-7 eV. As shown in Fig. 1, the simultaneous observation of both the Auger and conversion electrons resulted in spectra that could be examined in extremely fine detail, including quantification of the absolute emission rates and assessments of the secondary effects of electron shake-off and shake-up. The model calculations agree reasonable well with experiment, although these experiments indicate that the model may under-estimate the Auger-electron yields.



Figure 1. Low-energy L Auger and K conversion electron spectrum from the EC decay of ¹²⁵I [11.6].

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2.12. Nuclear Data Studies at IFIN-HH: Radionuclides for Medical Applications,

A. Luca

Scientific collaboration between researchers from IFIN-HH (Romania) and the IAEA-NDS, in the field of nuclear data has been long associated with nuclear structure and decay data evaluations, nuclear reaction data evaluations and cross section measurements, organization of training workshops and conferences. Recent evaluations of nuclear decay data for several radionuclides have been performed as part of the agreed work programmes for two IAEA CRPs: "Updated Decay Data Library for Actinides", 2005-2010 (²³⁶U, ²³⁴Th, ²²⁸Ra, ²¹¹Bi and ²¹¹Po), and "Nuclear Data for Charged-particle Monitor Reactions and Medical Isotope Production", 2012-2017 (⁵²Fe, ^{52m}Mn, ⁵²Mn, ²³⁰U and ²²⁶Th). Measurements of nuclear decay data (half-life and photon emission intensities) for radionuclides of medical interest have also been performed at IFIN-HH/DRMR, Radionuclide Metrology Laboratory (LMR) for ⁶⁴Cu, ⁶⁸Ga, ⁸⁹Zr, ¹²⁴I, ¹⁷⁷Lu and ¹⁸⁶Re.

A new TR19-type cyclotron and research laboratories into the production of radionuclides for medical applications are operational within the Radiopharmaceuticals Research Center (CCR) of IFIN-HH. While specific requests for improved nuclear decay data and cyclotron production reactions have been formulated, many of these needs have already been addressed and solved by previous IAEA-CRPs; various results are already in press or accepted for publication within Nucl. Data Sheets and J. Radioanal. Nucl. Chem. A particular interest of IFIN-HH was related to nuclear data for long-lived impurities produced in the $^{100}Mo(p,2n)^{99m}Tc$ reaction (alternative route for the production of ^{99m}Tc rather than the more common fission-based $^{99}Mo(\beta^{-})^{99m}Tc$ generator).

The Extreme Light Infrastructure – Nuclear Physics (ELI-NP) facility will be inaugurated in 2019 (www.eli-np.ro). Powerful advanced lasers and an intense high-energy gamma-ray beam will provide opportunities for new experimental research to produce radionuclides to develop existing and future procedures in nuclear medicine.

2.13. Cyclotron Production of ⁴⁷Sc as a Paradigm for Nuclear Data Needs, L. Canton

Co-workers: A. Fontana, M. Carante and S. Calzaferri (INFN-Pavia, Italy)

We have calculated the production of theranostic ⁴⁷Sc by proton collision with targets of ^{nat}Ti, ⁴⁸Ti, ⁴⁹Ti, ⁵⁰Ti and ^{nat}V. Analyses have been performed with the TALYS 1.9, EMPIRE 3.2 and FLUKA dev-2018.0 nuclear reaction codes. The main co-produced contaminant was found to be ⁴⁶Sc, which needs to be minimized in the irradiation process.

Both ^{nat}Ti and ⁴⁸Ti targets appear to be unsuitable for the production of high-purity ⁴⁷Sc at proton-beam energies below 100 MeV, while ⁴⁹Ti, ⁵⁰Ti and ^{nat}V targets exhibit appropriate energy regions for the production of theranostic ⁴⁷Sc: selected regions for optimum production are 25-40 MeV for enriched ⁴⁹Ti targets, 10-20 MeV for enriched ⁵⁰Ti targets, and 20-30 MeV for ^{nat}V targets.

We have calculated the activity produced from natural vanadium targets assuming 300 μ A proton beam with an incident energy of 29 MeV and outgoing energy of 24 MeV, corresponding to a target thickness of 600 μ m. Variations in both the pre-equilibrium model (pre-eqmodel range 1-4) and level-density model (ldmodel range 1-6) within the TALYS calculations provide a band of results that extend from maximum to minimum yields. Assuming 1 h irradiation, a maximum of 1.04 GBq and minimum of 56.32 MBq of ⁴⁷Sc are obtained at EOB, with the co-production of ⁴⁶Sc ranging from 50.69 kBq (max) to 8.28 kBq (min). Isotopic purity was maintained above 99.75% for the first 10 h

after EOB, and radionuclidic purity above 99% over the first three weeks. These results appear very promising and should stimulate TTY measurements in the corresponding energy; such measurements are planned within the Legnaro-INFN PASTA experiment (G. Pupillo, L. Mou, *et al.* – see Subsection 2.7, above).

As far as nuclear data needs are concerned, suitable ${}^{49}\text{Ti}(p,x){}^{47}\text{Sc}$ cross sections have yet to be measured, while the related ${}^{49}\text{Ti}(p,x){}^{46}\text{Sc}$ reaction to co-produce the main contaminant has only been measured once, although not in the relevant energy range (25-40 MeV). Moreover, there is only one measurement of the ${}^{50}\text{Ti}(p,x){}^{47}\text{Sc}$ and ${}^{50}\text{Ti}(p,x){}^{46}\text{Sc}$ production cross sections, which if possible needs to be repeated with better energy resolution.

Finally, while many data sets exist for ⁴⁷Sc production from ^{nat}V, a critical aspect for high-purity production is played by the threshold production of ⁴⁶Sc. The three nuclear reaction codes give significant differences in ⁴⁶Sc production in the energy region from 15 to 29 MeV which affect predictions of purity. Thus, precise measurements are required on the threshold production of ⁴⁶Sc in order to determine the purity levels that can be achieved.

2.14. A Medical Isotope App Based on TENDL, A.J. Koning, M. Verpelli and N. Gaughan

Nuclear Data Section staff are in the process of developing a web application for medical isotope production that will be based upon user input. As envisaged, the first version will be for incident charged particles only, while photonuclear reactions and neutron sources from e.g., research reactors will follow in a second stage.

The user specifies the characteristics of the incident particle source of interest:

- energy of the incident charged particles (e.g., protons),
- thickness of the target (or energy at the back of the target),
- power of the accelerator, and
- optional irradiation time,

after which the software returns, virtually instantaneously:

- yield of desired isotope as a function of time and/or energy window, and
- all impurities,

in a visually attractive and descriptive manner.

The current version of the graphical tool is based on:

- TENDL nuclear data library which provides complete nuclear data files for all incident particles, and
- ISOTOPIA: simple Fortran program which combines the TENDL library, analytical solutions for the basic production pathways (often sufficient), a radioactive decay data library, and a formula for the stopping power.

The demonstrated software shows various buttons to set the characteristics of the accelerator, the target isotope and the produced radioisotope, and a dynamical variation of production yields as a function of these variables. Suggested future improvements include the following:

- adoption of IAEA medical isotope database into TENDL to contribute towards a complete and good quality library,
- complete flexibility of the abundance of the target, including impurities for recycled targets, and
- adoption of more appropriate nomenclature for the various quantities.

A new version of the App will be released in due time to a few volunteers for them to test and provide user feedback with the aim of achieving further improvements through new additions and modifications.

3. DISCUSSIONS AND IDENTIFIED REQUIREMENTS

The basis of the initial discussions was dependent upon immediate assessments of various atomic and nuclear data needs that would or might arise during the course of the next five to twenty years with respect to immediate medical applications, clinical and pre-clinical trials and future on-going research-based developments. Data to be covered should include external beam radiotherapy (proton and hadron beams), and nuclear medicine (radionuclide production, targeted therapy) to be considered separately in terms of the decay data and excitation functions for optimum radionuclidic production:

- External beam therapy
 - a. hadron beams (Z>1),
 - b. proton beams.
 - Nuclear medicine

•

- a. application-related atomic and nuclear decay data, and more specifically the known need to improve the quantification of various Auger-electron and positron emission probabilities/intensities,
- b. radionuclide production
 - i. reaction cross sections,
 - ii. thick target yields (or "not thin" target yields).

3.1. External Beam Therapy

Perceived priority is higher for proton-beam therapy (more commonly used), high for carbon beams in current use, and lower for prospective beams (e.g., ⁴He beams).

3.1.1. Proton-beam therapy

- Gamma production cross sections for proton-induced reactions are required, especially for ¹²C,¹⁴N,¹⁶O(p,x γ) reactions, and perhaps also for other targets (e.g., ^{nat}P and ^{nat}Ca). Evaluations of measured data are merited for proton-beam energies < 85 MeV, along with the need for new measurements up to 250 MeV. Integral and energy differential (incident proton and emitted photon) data on prompt γ s up to μ s have been requested, as well as cross sections and branching ratios.
- Cross sections for reactions to produce positron emitters were defined as important: ¹²C(p,x)¹¹C, ¹⁴N(p,x)¹¹C and ¹⁶O(p,x)¹¹C; ¹⁴N(p,x)¹³N and ¹⁶O(p,x)¹³N; and ¹⁶O(p,x)¹⁵O, all for proton-beam energies up to 250 MeV. Cross-section data are also required that describe the reactions to produce ¹⁰C, especially ¹¹C(p,x)¹⁰C, and ¹²N, especially ¹⁴N(p,x)¹²N. Other reactions may also be significant, e.g., on targets of ¹⁶O, while noteworthy needs can also be identified with the ^{nat}P(p,x) and ^{nat}Ca(p,x) reactions.
- Target fragmentation as a function of beam energy and angle for proton-induced reactions were highlighted for necessary study, and some of these parameters will be measured via the FOOT (FragmentatiOn Of Target) collaboration.

3.1.2. Carbon beams

Production of ¹⁰C, ¹¹C, ¹³N, ¹⁵O positron emitters up to 200 MeV/nucleon; all data on ¹²C and ¹⁶O targets or fragmentation of the primary beam to produce positron-emitting residuals or heavy recoil projectiles (⁸B, ¹⁰C, ¹¹C).

3.1.3. ⁴He *beams*

Improved cross sections are required to produce ¹¹C, ¹³N and ¹⁵O positron emitters up to 200 MeV/nucleon.

More accurate data have been requested on projectile fragmentation as a function of energy and angle at therapy energies (200 MeV/nucleon) and up to ~ GeV energies for radiography.

3.1.4. Other relevant statements

Neutron production by proton interaction on light elements (such as Al) was judged to be an important requirement for spacecraft and glass in microscopes.

Possibility of future radioactive beams was noted, and requests for data on the interaction of, e.g., ¹¹C or ¹⁵O with physiological targets, can be anticipated.

Advances in accelerator design have reduced the interaction of the primary beams with collimators and other accelerator components; while the quantification of these reactions and neutron production are currently deficient, such data are expected to become increasingly irrelevant as a consequence of these continued developments.

3.2. Nuclear Medicine

3.2.1. Atomic and nuclear decay data

After some debate concerning the experience of evaluating specific decay-scheme data in detail during the course of the recent coordinated research project (2012-2017), agreement was reached that a less onerous and taxing start to such work would best be adopted in order to focus subsequently on important new decay-data measurements. Preliminary assessments of the existing atomic and nuclear decay data of each selected radionuclide should first be carried out, and any clear requirements for new measurements undertaken before an in-depth evaluation of the decay processes in order to recommend an improved and more substantive decay scheme.

A more practical route to obtaining an improved set of supportive decay data was suggested in the form of the following:

- very first step should always be to assess each individual decay scheme to be found within ENSDF; this exercise should include
 - relate these ENSDF decay data to later relevant papers,
 - assess and quantify fairly common gs to gs issues,
 - identify other decay issues,
 - other issues?
 - announce whether definitive new/improved measurement(s) are required;
- ensure these new required/requested measurements are undertaken;
- after all known new measurements have been published, consider requirement(s) for new evaluation(s);

• undertake a comprehensive decay-scheme evaluation for each radionuclide that satisfies the agreed criteria given above.

Requirements for improved decay data were identified/proposed and tabulated in specific presentations made during the course of Section 2 of the agenda (see specifically the Powerpoint presentations relevant to Sections 2.2 and 2.6), and these issues were considered further and re-defined during the course of this technical meeting. A listing of possible and known inadequacies and hence justifiably required decay data is given in Table 1, as agreed during the course of these extensive discussions.

Radionuclide	Decay data	Requirements
β⁺ emitters		
high priority:		
⁶¹ Cu, ⁷² As, ⁷⁴ As, ⁸⁶ Y	all β+, γ	assess need for measurements - if
		appropriate, measure and then fully evaluate
lower priority:		
⁴³ Sc, ⁵¹ Mn, ⁵⁷ Ni, ⁶⁶ Ga,	all β^+ , γ	assess need for measurements - if
⁷⁶ Br, ⁸¹ Rb, ^{82m} Rb, ⁸³ Sr,		appropriate, measure and then fully evaluate;
95 Ru, 120g I, 121 I, 134 La,		¹³⁵ La and ¹³⁵ Ce defined only as impurities in
$(^{135}La), ^{134}Ce, (^{135}Ce),$		¹³⁴ La and ¹³⁴ Ce, respectively
¹⁴⁰ Pr, ¹⁵² Tb	⁵¹ Mn (T _{1/2})	assess need for measurements
<u>β− emitters</u>		
high priority:		
⁴⁷ Sc	all β ⁻ , γ	assess need for measurements - if
		appropriate, measure and then fully evaluate;
⁶⁷ Cu, ¹⁸⁶ Re	all β ⁻ , γ	re-assess need for measurements - if
		appropriate, measure and then fully evaluate
lower priority:		
⁷⁷ As, ¹⁶¹ Tb, ¹⁷⁵ Yb	all β ⁻ , γ	assess need for measurements - if
		appropriate, measure and then fully evaluate
<u>a emitters</u>		
²²⁷ Th	all α, γ	assess need for measurements - if
		appropriate, measure and then fully evaluate
¹⁴⁹ Tb	all α , γ , β^+	re-assess need for measurements - if
		appropriate, measure and then fully evaluate
<u>diagnostic γ emitters</u>		
¹⁴⁷ Gd	all γ	assess need for measurements - if
		appropriate, measure and then fully evaluate
¹⁵⁵ Tb	all γ	assess need for measurements - if
		appropriate, measure and then fully evaluate
Auger (Ae) - and		
<u>conversion-electron</u>		
<u>(ce) emitters</u>		
high priority:		
^{117m} Sn	117m Sn (ce)	measurements and comparative modelling
		calculations required
¹²³ I, ^{193m} Pt, ^{195m} Pt,	123 I (Auger e),	measurements and comparative modelling
^{197m+g} Hg	^{193m,195m} Pt (Auger e),	calculations required
	^{197m+g} Hg (Auger e)	
lower priority:		
⁶⁷ Ga, ⁷¹ Ge, ⁷⁷ Br, ^{80m} Br,	Auger electrons and	CRP studies available for ^{103m} Rh and ¹¹¹ In;
^{103m} Rh, ¹¹¹ In, ¹¹⁹ Sb,	X-rays	others: require measurements and
¹³⁵ La, ¹⁶⁵ Er, ¹⁶⁹ Er		comparative modelling calculations

 Table 1. Compilation of Atomic and Nuclear Decay Data Requirements, December 2018.

Positron emitters:

High Priority: ${}^{61}Cu$, ${}^{72}As$, ${}^{74}As$ (longer T_{1/2} than ${}^{72}As$), ${}^{86}Y$.

Lower Priority: ⁴³Sc (positron intensity), ⁵¹Mn (assess half-life, and require new measurements), ⁵⁷Ni, ⁶⁶Ga, ⁷⁶Br (measured at FZJ, Julich ~ 12 years ago and re-evaluated 2017; complex decay scheme needs to be re-assessed), ⁸¹Rb, ^{82m}Rb, ⁸³Sr (available data inaccurate – new measurements required), ⁹⁵Ru, ¹²⁰gI (measured at FZJ, Julich ~ 12 years ago and re-evaluated 2017, but new measurements still needed), ¹²¹I, ¹³⁴Ce/¹³⁴La, (¹³⁴Ce/¹³⁴La constitute a potential *in vivo* generator, and ¹³⁵Ce/¹³⁵La are possible radioisotopic impurities), ¹⁴⁰Pr, ¹⁵²Tb (one of a theranostic pair).

Beta emitters:

High Priority: ⁴⁷Sc, ⁶⁷Cu (measured and re-evaluated in recent CRP – re-assess on basis of importance), ¹⁸⁶Re (re-assess on basis of importance); *Lower Priority:* ⁷⁷As, ¹⁶¹Tb, ¹⁷⁵Yb.

Alpha emitters:

¹⁴⁹Tb (re-assess need for measurements – if appropriate, measure and then fully evaluate).

Diagnostic gamma emitters:

¹⁴⁷Gd (assess decay data to determine priority (MRI+SPECT));

¹⁵⁵Tb (re-assess need for measurements – if appropriate, measure and then fully evaluate).

<u>Auger- and conversion-electron emitters</u> (also need to consider undertaking nuclear structure and decay evaluations as input for Auger-electron calculations):

High Priority: ^{117m}Sn (conversion electrons), ¹²³I, ^{193m}Pt, ^{195m}Pt, ^{197m+g}Hg (40 electrons per decay from ^{197m+g}Hg);

Lower Priority: ⁶⁷Ga, ⁷¹Ge ("pure" Auger-electron emitter), ⁷⁷Br, ^{80m}Br, ¹¹¹In (CRP study also completed recently), ¹¹⁹Sb, ¹³⁵La, ¹⁶⁵Er, ¹⁶⁹Er; possibly include ^{103m}Rh as part of any consideration of ¹⁰³Pd – but see also CRP 2012-2017.

Additional issues and requests:

Additional γ -ray measurements and structure and decay data evaluation for ¹²³I (both as γ emitter, and potential Auger-electron radiotherapy) and ^{149,152,155,161}Tb.

 99m Tc (P_{Ae}), 103 Pd (P_X, P_{Ae}), 111 In (P_{Ae}) were re-evaluated in the last CRP (to be published) to meet decay data needs and as input for Auger-electron calculations.

Auger- and electron-spectroscopic measurements are required to confirm the predictions of the BrIccEmis code. ^{99m}Tc, ¹⁰³Pd and ¹¹¹In atomic and nuclear decay data were evaluated as part of the previous CRP, and these results were used to benchmark the code. Such available experimental data are not presently compiled in any single location; these data need to be fully collected and assembled for use in the development of the code. Measurements should be made of other radionuclides that possess characteristic and dominant electron emissions, and comparable code calculations should be disseminated directly by the IAEA as a result of future evaluations. Further improvements and additional developments to the codes are also to be encouraged.

3.2.2. Cross-section production data

Specific needs of prompt-gamma production cross sections were highlighted for online range (dose) verification in proton- and heavy-ion radiotherapy: Review/assess prompt-gamma production cross sections from nuclear de-excitation on biological targets ^{12,13}C, ^{15,14}N, ¹⁶⁻¹⁸O and/or heavy-ion beam fragments ¹¹C, ¹³N, ¹⁵O to be used in proton and heavy-ion range verification studies leading to online verification systems (e.g., see

"Prompt gamma detection for range verification in proton therapy", S. Kurosawa, H. Kubo, S. Iwaki, et al, *Curr. Appl. Phys.* **12** (2012) 364-368).

Specific reactions with relevance to charged-particle beam monitoring, SPECT and PET radionuclide generation, and the production of radionuclides with therapeutic relevance were identified and are individually listed below. As in previous CRPs, consideration of radionuclidic impurities in the production of radionuclides of interest is considered relevant, whether investigated experimentally or predicted by means of theoretical codes. Previously evaluated reactions have also been reconsidered if new data are available, along with other specific areas of general importance:

Many reactions are in need of measurement in the "intermediate" energy region between 40 and approximately 200 MeV, which is within the energy range of several modern radionuclide production facilities. Monitor reaction data in this energy range that are free from secondary neutron contamination, e.g., (p,xn)-type reactions, are of significant importance. Several reactions previously evaluated require upward energy extension to 200 MeV, for example: ⁵⁵Mn(p,4n)⁵²Fe, ⁵⁹Co(p,3n)⁵⁷Ni, ⁶⁸Zn(p,*a*n)⁶⁴Cu, ⁷¹Ga(p,4n)⁶⁸Ge, ⁷⁵As(p,3n)⁷³Se, ⁸⁵Rb(p,3n)⁸³Sr, ⁸⁸Sr(p,3n)⁸⁶Y, ¹²⁵Te(p,2n)¹²⁴Te, ¹²⁴Xe(p,pn)¹²³Xe, ¹²⁴Xe(p,2p)¹²³I, ⁴⁵Sc(p,2n)⁴⁴Ti, ⁶⁹Ga(p,2n)⁶⁸Ge, ^{nat}Br(p,x)⁷²Se, ⁶⁸Zn(p,2p)⁶⁷Cu, and ²³²Th(p,x)²²⁵Ac. These reactions may be considered of lower priority than those discussed explicitly below.

More especially (p,n) along with some other reactions require careful measurement or theoretical adjustment in the energy region near threshold where the cross section is most rapidly increasing with energy. These measurements are made more important by the proliferation of low-energy facilities for the production of radionuclides through solution target bombardment, self-shielded systems, and purpose-driven machines, e.g., for ¹⁵O production.

Neutron-induced reactions from high-energy spallation neutrons also require supporting data, with needs present up to 200-MeV incident energy. Several high-energy monitor reactions are in need of additional measurements (especially multiple neutron emission reactions on Bi, Lu and Tm) up to 200-MeV neutron energy. These data are being considered by the neutron dosimetry community for high-energy applications. Reactions of higher priority with relevance to medical radionuclide production include 232 Th(n,x)^{225,227}Ac, 70 Zn(n, α)⁶⁷Ni, 50 Ti(n,x)⁴⁷Sc, 50 Ti(n, α)⁴⁷Ca, and 226 Ra(n,2n)²²⁵Ra.

The validation of data by yield measurements is important for such features as the careful selection of experimental energy windows, precise experimental design and thorough documentation. An alternative, simpler method of monitor reaction validation that has been suggested is to quantify multiple well-characterized reactions in the same irradiation, in order to confirm agreement with fluxes quantified by means of the various reactions in a single foil/target (e.g., Ti, Cu).

Where alpha beams are available, emphasis should be placed on the exploration of reactions which produce high-spin isomers of radionuclides with medical importance. There is particular interest in the potential of future radioactive beams with relevance to physiological targets, e.g., of ¹¹C or ¹⁵O.

Photonuclear production methods are being investigated and seen as increasingly viable, although they are supported by rather sparse data. Reactions that take place in bremsstrahlung converters are also of interest. Compilations of such reactions are maintained both by the US DOE and an IAEA CRP on photonuclear reactions (based on an IAEA CRP from 2000).

Specific reactions that have not been evaluated before, but need consideration are as follows:

Monitor Reactions

Proton-induced reactions:

 $^{27}Al(p,x)^7Be$, $^{nat}Ni(p,x)^{56}Ni$, $^{nat}Ni(p,x)^{57}Ni$, $^{nat}Cu(p,xn)^{62/65}Zn$, $^{93}Nb(p,4n)^{90}Mo$, $^{197}Au(p,pn)^{196g}Au$, $^{197}Au(p,p3n)^{194}Au$, $^{197}Au(p,n)^{197m+197g}Hg$ **Deuteron-induced reactions**:

 $^{nat}Ni(d,x)^{57}Ni$, $^{197}Au(d,x)^{196g}Au$, $^{197}Au(d,x)^{194}Au$, $^{197}Au(d,x)^{198}Au$, $^{197}Au(d,x)^{197m/197g}Hg$

Radionuclides for Single Photon Emission Computed Tomography (SPECT)

⁹⁹mTc. Assessment of radioisotopic impurities produced from all relevant target Mo isotopes should be attempted.

⁹⁷**Ru.** ${}^{103}Rh(p,x){}^{97}Ru$, ${}^{99}Tc(p,3n){}^{97}Ru{}^{(6)}$, ${}^{nat}Mo(\alpha,x){}^{97}Ru$. ${}^{97}Ru$ forms a theranostic pair SPECT with ${}^{103}Ru$; compounds formed with Ru are more stable than those with ${}^{99m}Tc$.

¹⁵⁵**Tb.** ¹⁵⁵ $Gd(p,n)^{155}Tb$, ¹⁵⁶ $Gd(p,2n)^{155}Tb$, and ¹⁵⁵ $Gd(d,2n)^{155}Tb$. ¹⁵⁵Tb is theranostic with ¹⁴⁹Tb and ¹⁶¹Tb.

¹⁹¹Os. ¹⁹⁰Os $(d,p)^{191}$ Os. Ultrashort-lived radionuclides offer several advantages in angiocardiography when compared with ^{99m}Tc. Of the isotopes proposed for this application, ^{191m}Ir (4.899 s) has several advantages when compared with ^{195m}Au (30.5 s) (longer generator shelf-life) and ^{109m}Ag (40 s) (higher photon yield).

Radionuclides for Positron Emission Tomography (PET)

³⁴mCl. ³⁶ $Ar(d,\alpha)^{34m}Cl$, ^{nat} $Cl(p,x)^{34m}Cl$, ³² $S(\alpha,pn)^{34m}Cl$, ³⁵ $Cl(p,pn)^{34m}Cl$, ^{nat} $S(p,n)^{34m}Cl$, ^{nat} $S(d,2n)^{34m}Cl$, ³¹ $P(\alpha,n)^{34m}Cl$. Only positron-emitting radioisotope of chlorine.

³⁸K. ³⁸Ar(p,n)³⁸K, ⁴⁰Ar(p,3n)³⁸K, ³⁵Cl(α ,n)³⁸K. Useful in nuclear cardiology.

⁴³Sc. ⁴³*Ca*(*p*,*n*)⁴³*Sc*, ⁴²*Ca*(*d*,*n*)⁴³*Sc*, ⁴⁰*Ca*(*a*,*p*)⁴³*Sc*, ⁴⁰*Ca*(*a*,*n*)⁴³*Sc*. Theranostic with β^- emitting ⁴⁷*Sc*. Emits fewer high-energy γ rays than ⁴⁴*Sc*.

⁴⁵Ti. ⁴⁵Sc(p,n)⁴⁵Ti, ⁴⁵Sc(d,2n)⁴⁵Ti. Can be produced in extremely high yields from targets of naturally monoisotopic scandium; has a lower positron energy than any other radionuclide apart from ¹⁸F.

⁵¹**Mn.** ⁵⁴ $Fe(p,\alpha)^{51}Mn$, ⁵⁴ $Fe(d,\alpha n)^{51}Mn$, ⁵⁰ $Cr(d,n)^{51}Mn$, ^{nat} $V({}^{3}He,x)^{51}Mn$, ^{nat} $Fe(p,x)^{51}Mn$. Short-lived, low-dose alternative to ^{52g}Mn (T_{1/2} = 5.59 d, β^+ = 29.6%), which has several high energy gamma rays.

⁶¹Cu. ^{*nat*} $Ni(p,x)^{61}Cu$. Previously evaluated data should be included for this reaction in the monitor reaction section of the IAEA-NDS nuclear data portal.

⁶⁹Ge. ^{*nat*}Ga(p,x)⁶⁹Ge, ^{*nat*}Ga(d,x)⁶⁹Ge, ⁶⁹Ga(p,x)⁶⁹Ge, ⁶⁹Ga(d,2n)⁶⁹Ge. Attractive PET isotope that is theranostic with ⁷¹Ge, a pure Auger-electron emitter.

⁷²Se/⁷²As. ^{*nat*}Rb(p,x)⁷²Se at high energy and ⁷⁵As(p,4n)⁷²Se (and production of ^{70,71,73,75}Se) up to 200 MeV. ⁷²As is PET-imaging counterpart for β -emitting ⁷⁷As.

⁷⁶Br. ⁷⁹Br $(p,4n)^{76}$ Kr $(14.8 h) \rightarrow ^{76}$ Br. Theranostic with Auger-electron emitter ⁷⁷Br.

⁸³Sr. ⁸⁵*Rb*(*p*,3*n*)⁸³Sr, ⁸²*Kr*(³*He*,2*n*)⁸³Sr. Theranostic with β⁻ emitter ⁸⁹Sr.

⁸⁶Y. ^{87g,87m,88}Y from ^{87,88}Sr up to 50 MeV. ⁸⁹Y(p,4n)⁸⁶Zr \rightarrow ⁸⁶Y. Theranostic with ⁸⁹Sr, and newly proposed for production via higher energy reactions on targets of ⁸⁹Y.

¹⁵²**Tb.** ${}^{155}Gd(p,4n)^{152}Tb$. Several potential theranostic applications when used with terbium radionuclides 149 Tb, 155 Tb, and 161 Tb.

Therapeutic Radionuclides

 $^{^{6}}$ Although mentioned in discussions at the meeting, this reaction with a radioactive target is unlikely to be used in the production of 97 Ru.

⁴⁶Sc. $^{nat}Ti(p,x)^{46}Sc$, $^{nat}Ti(d,x)^{46}Sc$, $^{45}Sc(d,p)^{46}Sc$, $^{45}Sc(^{3}He, 2p)^{46}Sc$, $^{45}Sc(a,2pn)^{46}Sc$. A radioisotopic impurity in productions of ⁴³Sc, ⁴⁴Sc, and ⁴⁷Sc.

⁴⁷Sc. ⁴⁸Ca(p,2n)⁴⁷Sc, ⁴⁸Ca(p,pn)⁴⁷Ca \rightarrow ⁴⁷Sc, ⁴⁸Ti(p,2p)⁴⁷Sc, ⁴⁹Ti(p,x)⁴⁷Sc, ⁴⁹Ti(p,3p)⁴⁷Ca \rightarrow ⁴⁷Sc, ⁵⁰Ti(p,x)⁴⁷Sc, ⁵⁰Ti(p,x)⁴⁷Ca \rightarrow ⁴⁷Sc, ^{nat}V(p,x)⁴⁷Sc, ⁴⁸Ca(y,x)⁴⁷Ca \rightarrow ⁴⁷Sc, ${}^{48}Ca(n,2n){}^{47}Ca \rightarrow {}^{47}Sc, \; {}^{46}Ca(n,\gamma){}^{47}Ca \rightarrow {}^{47}Sc, \; {}^{48}Ca(p,2n){}^{47}Sc, \; {}^{48}Ca(d,3n) \; {}^{47}Sc. \; {}^{47}Sc$ is the therapeutic radioisotope of scandium that is combined with positron-emitting diagnostic radioisotopes ⁴³Sc and ⁴⁴Sc, and produced with radioisotopic impurities ⁴⁶Sc and ⁴⁸Sc. All possible routes to ⁴⁷Sc should be evaluated.

^{58m}Co. (lower priority). ${}^{57}Fe(d,n){}^{58m}Co, {}^{58}Fe(p,n){}^{58m}Co.$ Targeted and Auger-electron based radioimmunotherapy and PET-imaging post-therapy with daughter ^{58g}Co.

⁶⁷Cu. ⁷⁰Zn(p,x)⁶⁷Cu, ⁶⁸Zn(p,2p)⁶⁷Cu. Theranostic with positron-emitting ⁶⁴Cu, ⁶²Cu and ⁶¹Cu. New measurements have been made, and all data should be evaluated up to 70 MeV. ⁷¹Ge. ^{nat}Ga(p,x)⁷¹Ge, ^{nat}Ga(d,x)⁷¹Ge, ⁷¹Ga(p,n)⁷¹Ge, ⁷¹Ga(d,2n)⁷¹Ge. Pure electroncapture decay, and potentially theranostic with ⁶⁹Ge. Also relevant to large-scale production of ⁶⁸Ge from gallium targets.

 $^{78}Se(p,2n)^{77}Br,$ ⁷⁷Br. $^{75}As(\alpha,2n)^{77}Br$, $^{77}Se(p,n)^{77}Br$, $^{79,81}Br(p,xn)^{77}Kr \rightarrow ^{77}Br$ $^{nat}Rb(p,x)^{77/82}Br$. Auger-electron emitter theranostic with positron-emitting ⁷⁶Br. Produced by low-energy reactions on selenium, and also from Rb targets irradiated at high energy to produce ⁸²Sr.

^{80m}Br. ⁸⁰Se $(p,n)^{80m}Br$, ^{nat}Se $(p,xn)^{80m}Br$, ⁸⁰Se $(d,2n)^{80m}Br$. Short-lived Auger-electron emitter theranostic with positron-emitting ⁷⁶Br.

 $^{232}Th(p,f)^{103,106}Ru, \quad {}^{100}Mo(\alpha,n)^{103}Ru,$ $^{102}Ru(n,\gamma)^{103}Ru,$ $^{104}Ru(\gamma,n)^{103}Ru,$ ¹⁰³Ru. ${}^{104}Ru(n,2n){}^{103}Ru$, ${}^{103}Rh(n,p){}^{103}Ru \rightarrow {}^{103m}Rh$. ${}^{103}Ru$ is the parent isotope of ${}^{103m}Rh$, an Auger-electron emitter.

¹⁰³Pd. ${}^{107}Ag(p,\alpha n){}^{103}Pd$. Auger emitter. Reactions that produce associated impurities up to 100 and preferably 200 MeV are also recommended for consideration.

 $^{121}Sb(p,2pxn)^{113,117m,121m,121g}Sn.$ ^{117m}Sn. $^{116}Cd(\alpha,3n)^{117m}Sn,$ $^{nat}In(\alpha, pxn)^{117m}Sn,$ $^{nat}Sb(p,x)^{113,117m,121m,121g}Sn$. Low-energy electron-emitting therapeutic radionuclide with own 159-keV y ray for SPECT.

 ${}^{119}Sb. {}^{119}Sn(p,n)^{119}Sb, {}^{119}Sn(d,2n)^{119}Sb, {}^{120}Sn(p,2n)^{119}Sb, {}^{121}Sb(p,3n)^{119}Te \rightarrow {}^{119}Sb, {}^{121}Sb(d,4n)^{119}Te \rightarrow {}^{119}Sb, {}^{122}Sn(p,4n)^{119}Sb, {}^{117}Sn(a,2n)^{119}Te \rightarrow {}^{119}Sb, {}^{117}Sn(a,2n)^{119}Te \rightarrow {}^{119}Sb, {}^{117}Sn(a,2n)^{119}Te \rightarrow {}^{119}Sb, {}^{119}Sb, {}^{117}Sn(a,2n)^{119}Te \rightarrow {}^{119}Sb, {}^{119}Sb, {}^{117}Sn(a,2n)^{119}Sb, {}^{119}Sb, {}^{117}Sn(a,2n)^{119}Sb, {}^{119}Sb, {}^{119}Sb$ $^{117}Sn(^{3}He,n)^{119}Te \rightarrow ^{119}Sb$. Potent Auger-electron emitter for therapy. Reactions to produce radioisotopic impurities of ^{117/118}Sb are also important.

¹³⁴Ce/¹³⁴La. $^{139}La(p,6n)^{134}Ce$ and reactions to produce main impurities of ^{132,133,135,137,139}Ce up to 200 MeV. Auger-electron and positron-emitting *in vivo* generator for radiotherapy. Positron-emitting ¹³⁴La can be used to study kinetics and dosimetry by means of PET.

¹³⁵La and ¹³²La. $^{nat}Ba(p,x)^{131,132,133,135,137}La$, $^{135}Ba(p,n)^{135}La$, $^{136}Ba(p,2n)^{135}La$. Augerelectron emitter (135 La) and positron emitter (132 La).

¹⁶¹**Tb.** ${}^{160}Gd(n,\gamma){}^{161}Gd \rightarrow {}^{161}Tb, {}^{160}Gd(d,n){}^{161}Tb.$ Combined β -/Auger-electron therapy. ¹⁶⁵**Er.** ${}^{165}Ho(p,n){}^{165}Er, {}^{165}Ho(d,2n){}^{165}Er, {}^{166}Er(p,2n){}^{165}Tm \rightarrow {}^{165}Er,$ $^{166}Er(d,3n)^{165}Tm \rightarrow ^{165}Er$, $^{nat}Er(p,xn)^{165}Tm \rightarrow ^{165}Er$ and $^{164}Er(d,n)^{165}Tm \rightarrow ^{165}Er$. Augerelectron emitter for therapy.

 $^{167}Er(d,2n)^{167}Tm$, $^{167}Er(p,n)^{167}Tm$, $^{165}Ho(\alpha,2n)^{167}Tm$, ¹⁶⁷Tm. $^{nat}Er(p,xn)^{167}Tm,$ $^{169}Tm(n,3n)^{167}Tm$. ^{167}Tm emits prominent 208-keV γ ray and low-energy electrons.

¹⁸⁶**Re.** ¹⁸⁹ $Os(p,x)^{186}Re. \beta^-$ emitter for radiotherapy. Excitation functions for production of co-produced impurities ^{181,182m,182g,183,184,184g,188m,188g,189}Re, as well as precursors such as ^{181,182,183}Os and ^{181,182,183}Ir should be considered up to a beam energy of 50 MeV. ¹⁸⁹Re. ¹⁹²Os(p, α)¹⁸⁹Re, ¹⁹²Os(d, α n)¹⁸⁹Re, ¹⁸⁶W(α ,p)¹⁸⁹Re. β^- radiotherapy primarily for

metastatic bone pain; also emits diagnostic 216.7-keV γ ray.

¹⁹⁸gAu. ¹⁹⁸Pt(d,2n)¹⁹⁸Au, ¹⁹⁸Pt(p,n)¹⁹⁸Au. β^- emission ideal in targeted radiotherapy applications, while emission of low-energy photon allows a simultaneous evaluation of dose distributions via SPECT imaging.

¹⁹⁷mHg and ¹⁹⁷gHg. ${}^{197}Au(p,3n)^{197m+g}Hg$, ${}^{197}Au(d,2n)^{197m+g}Hg$. Both ${}^{197m}Hg$ and ${}^{197g}Hg$ are candidates for Auger-electron therapy.

²²⁵Ac. ²²⁶Ra(n,2n)²²⁵Ra \rightarrow ²²⁵Ac, ²²⁶Ra(γ ,x)²²⁵Ra \rightarrow ²²⁵Ac, ²²⁶Ra(p,2n)²²⁵Ac, ²²⁶Ra(p,n)²²⁶Ac, ²²⁶Ra(n,x)²²⁹Th, ²³²Th(p,x)²²⁹Th, ²³²Th(p,x)²²⁵Ra \rightarrow ²²⁵Ac. ²²⁵Ac is among the most promising radionuclides for targeted alpha therapy. New measurements are urgently required to resolve discrepancies in previously evaluated datasets.

²³⁰Pa \rightarrow ²³⁰U \rightarrow ²²⁶Th. ²³²Th(p,3n)²³⁰Pa, ²³²Th(d,4n)²³⁰Pa. A decay chain with multiple potential radionuclides for targeted alpha therapy, especially ²³⁰U and ²²⁶Th. Previously evaluated, but in need of new measurements to resolve discrepancies in existing datasets.

No measurements of the energy differential excitation function are planned, but reactions induced by fast neutrons are of interest to radionuclide production. If an incident neutron spectrum is provided, a future CRP could evaluate the reaction, which would be especially relevant to the production of ^{90,90m}Y, ⁶⁷Cu, ⁴⁸Sc, and/or ³²P that all proceed by (n,p)-type reactions and can be induced by breakup deuterons from D-D and D-T sources.

4. CONCLUSIONS

The content of this report contains a subjective selection of relevant radionuclides based on our knowledge of existing and emerging developments within external particle beam therapy and diagnostic and therapeutic nuclear medicine. Participants assessed and reviewed the proposed requirements for cross-section and decay data of a significant number of radionuclides deemed as suitable for such medical applications. Recommendations focused on the need for cross-section studies over a reasonably wide range of targets and projectiles, along with atomic and nuclear decay data measurements for specific radionuclides, as described in Section 3. Further thought also needs to be given to measuring and evaluating the equivalent data for various radionuclidic impurities that may also be produced during the irradiations of interest. The need to provide the user community with uncertainty values in the recommended data was also noted. 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 diagnostic and therapeutic treatments and relevant research studies.

Consideration of external beam therapy by means of protons and heavier ions focused on highpriority needs to verify and validate the delivery of the planned dose for both protons and heavy ions. In particular, gamma production cross sections for proton-induced reactions are required, especially for ¹²C, ¹⁴N, ¹⁶O(p,x γ) reactions, and activation products relevant to proton and heavyion radiotherapy need to be better quantified up to 250 MeV/nucleon with respect to their production cross sections (particularly the positron-emitters ^{10,11}C, ^{12,13}N, ¹⁵O, ³⁰P and ³⁸K). Evaluations of measured data are merited for proton-beam energies < 85 MeV, along with the need for new measurements up to 250 MeV. Target fragmentation as a function of beam energy and angle for proton-induced reactions were also highlighted for necessary study.

Much has happened involving progress in measurements and evaluations of various atomic and nuclear decay data since the previous IAEA-NDS exercise of August 2011 to define at that time present and future data requirements for diagnostic and therapeutic applications in nuclear medicine. As stated earlier, these previous recommendations were directly but only partially

addressed during the course of 2012 to 2017 by means of an IAEA coordinated research project. Therefore, further re-assessments have been undertaken in December 2018 with respect to the remaining balance of outstanding needs from August 2011, along with consideration of other relevant in-depth studies that have appeared in the intervening years.

5. RECOMMENDATIONS

The newly updated requirements for improved decay data are listed in Table 1, and summarised more briefly below on the basis of an initial comprehensive need for such a re-assessment of the existing data for each radionuclide, possibly leading on to important new measurements prior to any attempted in-depth re-evaluation of the full decay scheme:

β^+ emitters

high priority ⁶¹Cu, ⁷²As, ⁷⁴As and ⁸⁶Y; lower priority (also impurities generated in their production) ⁴³Sc, ⁵¹Mn, ⁵⁷Ni, ⁶⁶Ga, ⁷⁶Br, ⁸¹Rb, ^{82m}Rb, ⁸³Sr, ⁹⁵Ru, ¹²⁰gI, ¹²¹I, ¹³⁴La, ¹³⁴Ce, ¹⁴⁰Pr and ¹⁵²Tb

β^- emitters

high priority ⁴⁷Sc, ⁶⁷Cu and ¹⁸⁶Re; lower priority ⁷⁷As, ¹⁶¹Tb and ¹⁷⁵Yb

α emitter ²²⁷Th and ¹⁴⁹Tb

γ emitters ¹⁴⁷Gd, ¹⁵²Tb, ¹⁵⁵Tb and ¹⁶¹Tb

Auger- and conversion-electron emitters

high priority ^{117m}Sn(ce), ¹²³I(Ae), ^{193m}Pt(Ae), ^{195m}Pt(Ae) and ^{197m+g}Hg(Ae) lower priority ⁶⁷Ga, ⁷¹Ge, ⁷⁷Br, ^{80m}Br, ^{103m}Rh, ¹¹¹In, ¹¹⁹Sb, ¹³⁵La, ¹⁶⁵Er and ¹⁶⁹Er

Some of these radionuclides have arisen as a consequence of work undertaken during the course of the recent CRP (⁶¹Cu, ⁶⁷Cu, ⁷⁶Br, ^{103m}Rh, ¹¹¹In and ^{120g}I), while many more constitute previously unaddressed and now defined as the re-assessed remainder of August 2011 along with significant new additions that have arisen from more recent sources of information.

Nuclear reaction data require additional evaluation in the context of charged-particle beam monitoring, especially for the following reactions: ${}^{27}Al(p,x)^7Be$, ${}^{197}Au(p,pn)^{196g}Au$, ${}^{197}Au(p,n)^{197m+g}Hg$, ${}^{197}Au(p,p3n)^{194}Au$, ${}^{nat}Ni(p,x)^{57}Ni$, ${}^{nat}Ni(p,x)^{56}Ni$, ${}^{93}Nb(p,4n)^{90}Mo$, ${}^{nat}Cu(p,xn)^{62,65}Zn$, ${}^{197}Au(d,x)^{196g}Au$, ${}^{197}Au(d,x)^{197m+g}Hg$, ${}^{197}Au(d,x)^{194}Au$, ${}^{197}Au(d,x)^{198}Au$, and ${}^{nat}Ni(d,x)^{57}Ni$.

Imaging applications with the following radionuclides should also be evaluated, along with the production of radionuclidic impurities generated in likely production schemes: ⁹⁷Ru, ¹⁵⁵Tb, ¹⁹¹Os, ^{99m}Tc, ³⁸K, ^{34m}Cl, ⁴³Sc, ⁴⁵Ti, ⁵¹Mn, ⁶¹Cu, ⁶⁹Ge, ⁷²Se/⁷²As, ⁷⁶Br, ⁸³Sr, ⁸⁶Sr and ¹⁵²Tb. Specific therapeutic radionuclides are of rapidly increasing interest in various medical fields, and should be evaluated (or re-evaluated, especially in cases where prior work has suggested that new measurements be made and they have been subsequently undertaken): ⁴⁷Sc, ⁴⁶Sc, ^{58m}Co, ⁷¹Ge, ⁶⁷Cu, ⁷⁷Br, ^{80m}Br, ¹⁰³Ru, ¹⁰³Pd, ^{117m}Sn, ¹¹⁹Sb, ¹³⁴Ce/¹³⁴La, ¹³⁵La and ¹³²La, ¹⁶¹Tb, ¹⁶⁷Tm, ¹⁶⁵Er, ¹⁸⁶Re, ¹⁸⁹Re, ^{197m}Hg + ^{197g}Hg, ^{198g}Au, ²²⁵Ac, ²³⁰U and ²²⁶Th. As these radionuclides are considered, the energy ranges near their reaction thresholds and in the intermediate region between 40 and 200 MeV should receive special consideration, since these data are commonly sparse.

Radionuclide production for medical applications is evolving towards the regular need for intermediate-energy, high-power accelerators with extended proton energies up to 250 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 is also a growing requirement for such facilities to generate deuterons, ³He and ⁴He particle beams in order to extend the range of production and improve radionuclidic purity.

Consideration should be given to the organization of joint IAEA-ICTP workshops on Nuclear Data for Medical Applications to be held on a regular basis (every five or six years). Emphasis should be placed on the atomic and nuclear data required to undertake efficacious diagnostic and therapeutic procedures that constitute the preferred methods of production and means of defining and quantifying patient dose.

An entirely adequate dissemination system has been developed by IAEA Nuclear Data Section staff to handle and display all recommended excitation functions and atomic and nuclear data that have arisen from any relevant data evaluation and assembly initiatives instigated by the IAEA. As part of this set of aims, the BrIccEmis code has been developed as a tool 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 previously available. The aim has always been to produce a definitive and consistent set of data of all the nuclides of immediate value to the medical profession. These resulting recommended sets ofcross-section and decay data can be found at

www-nds.iaea.org/medportal/

and strong emphasis should be placed on maintaining, extending and improving these datasets at regular intervals into the future.

The IAEA is urged to consider the recommendations contained within this report, as brought together on the basis of the agreed atomic and nuclear data needs for medical radionuclides over the next 5 to 10 years. Some modest attempt was made to prioritize the various data requirements identified as important over the specified timescale stretching towards 2035/2040. Plans for appropriate work programmes to develop and improve such atomic and nuclear data over this same timescale should involve serious note being taken of the requirements and recommendations contained in detail throughout Section 3, above.

APPROVED AGENDA

Technical Meeting on Nuclear Data for Medical Applications

IAEA, Vienna, Austria 10-13 December 2018 Meeting Room VIC MOE03

APPROVED AGENDA

Monday, 10 December

- 09:00 09:30 Registration (IAEA Registration Desk, Gate 1)
- **09:30 10:30 Opening Session**

Welcoming address (Arjan Koning (SH-NDS)) Administrative matters Election of Chairman and Rapporteurs Adoption of the Agenda

Overview of Evaluated Cross Sections for Medical Isotope Production Within IAEA CRPs, R. Capote Noy

10:30 – 17:30 Presentations by participants (~ 30 min each)

Monitor Reactions: CRP Input, Outcome and Lessons Learned During 2014-2018, A. Hermanne

Atomic and Nuclear Decay Data for Medical Applications, A.L. Nichols

Nuclear Data Needs for Medical Applications Using Deuteron Breakup Neutrons, Y. Nagai

Addressing Nuclear Data Needs of the US DOE Isotope Program, F.M. Nortier

Novel Medical Radionuclides, QMN Neutrons, and Thick Target Yields, J.W. Engle

Development of Novel Radionuclides for Medical Applications, S.M. Qaim

Coffee break(s) as needed

(12:30 – 14:00 Lunch break)

Tuesday, 11 December

09:30 - 17:30	Presentations by participants (cont'd)
	Cross-section Measurements of Proton-induced Reactions for the Production of ⁶⁷ Cu and ⁴⁷ Sc, G. Pupillo and L. Mou
	Nuclear Data Needs Related to Hadron Therapy, A. Ferrari
	Argentine Project to Develop the Production of ²²⁵ Ac, ²¹³ Bi and Suitable Radiopharmaceuticals for Therapy: Nuclear Data Needs, S. Siri
	Measurements of Production Yields of Positron Emitters Relevant for Beam Range Verification in Proton Therapy, C. Guerrero
	New Measurements of the Absolute Auger-electron Yields of Medical Radionuclides, and Recent Developments of the BrIccEmis Code, T. Kibédi
	Nuclear Decay Data Studies at IFIN-HH: Radionuclides for Medical Applications, A. Luca
	<i>Cyclotron Production of</i> ⁴⁷ <i>Sc as a Paradigm for Nuclear Data Needs</i> , L. Canton
	A Medical Isotope App Based on TENDL, A.J. Koning, M. Verpelli and N. Gaughan
	Coffee break(s) as needed

(12:30 – 14:00 Lunch break)

19:00 Dinner at a restaurant (see separate information in the folder)

Wednesday, 12 December

scussion	
nary Report	
unch break)	Coffee break(s) as needed
	nch break)

Thursday, 13 December

09:30 - 15:00	Drafting of Summary Report (cont'd)	
	Closing of the Meeting	
	(12:30 – 14:00 Lunch break)	Coffee break(s) as needed

Technical Meeting on Nuclear Data for Medical Applications 10 to 13 December 2018 IAEA, Vienna

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APPENDIX 3

Technical Meeting Nuclear Data Needs for Medical Applications

MEETING PHOTOGRAPH



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