Memo CP-D/164

9 October 1987

To: Distribution

From: K. Okamoto and H.D. Lemmel

Subject: Data for medical radioisotope production

Please find attached the Conclusions and Recommendations of three working groups of the IAEA Consultants' Meeting on Data Requirements for Medical Radioisotope Production, Tokyo, 20-24 April 1984.

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IAEA Consultants' Meeting on
DATA REQUIREMENTS FOR MEDICAL RADIOISOTOPE PRODUCTION

in co-operation with the
Institute of Physical and Chemical Research (RIKEN)
Tokyo, 20-24 April 1987

SUMMARY OF CONCLUSIONS AND RECOMMENDATIONS

Working Group I: EXPERIMENTAL DATA

R.M. Lambrecht (Chairman), S.L. Waters (Co-chairman), Lu Hanlin, S.M. Qaim, H. Umezawa, G.J. Beyer, H. Heinzl, M. Bonardi, T. Nozaki, A. Hashizume, M.C. Lagunas-Solar, K. Kitao, D. Berényi

The standardization of reported nuclear data for medical radioisotope (MRI) production could serve to, (i) establish and maintain international uniformity; (ii) improve the accuracy, where this would become necessary; and (iii) help developing laboratories and hospital- based medical cyclotron facilities. It was felt that there is a need to standardize the following parameters

- The type of particle accelerator and the incident beam energy and resolution set by the magnetic field or as measured by either time-of-flight (neutron and/or gamma flash) or monitor reactions should always be reported.

  It was concluded that monitor reactions would be the most convenient and important to the largest number of laboratories involved in MRI production. The errors in incident energy, energy degradation in the target and the estimated error in stopping power should be cited.

- The following monitor reactions are very useful for beam energy and intensity measurements. These reactions were chosen on the basis of reported cross sections, decay properties of product nuclei and the suitability of target materials as monitor foils.

$^{12}C(p,pn)^{11}C$
$^{27}Al(p,3pn)^{24}Na$
$^{27}Al(p,3p3n)^{22}Na$
$^{59}Co(p,pn)^{58}Co$
$^{63}Cu(p,n)^{63}Zn$
$^{63}Cu(p,2n)^{62}Zn$
$^{65}Cu(p,n)^{65}Zn$
$^{nat}Cu(p,x)^{61}Cu$
$^{27}Al(d,3p2n)^{24}Na$
$^{51}V(d,2n)^{51}Cr$
$^{56}Fe(d,n)^{57}Co$
$^{nat}Ti(^{3}He,x)^{48}V$
$^{63}Cu(^{3}He,p3n)^{62}Zn$
$^{65}Cu(^{3}He,2n)^{66}Ga$
$^{65}Cu(^{3}He,p2n)^{65}Zn$

$^{27}Al(^{4}He,4p3n)^{24}Na$
$^{63}Cu(^{4}He,n)^{66}Ga$
$^{63}Cu(^{4}He,pn)^{65}Zn$
$^{65}Cu(^{4}He,2n)^{67}Ga$

- It was recognized that there is no general agreement on the use of standard reactions for monitoring beam energy and intensities and that this may be responsible for some of the discrepancies in the reported cross section data. It was strongly recommended that the IAEA make arrangements at the Agency or at another nuclear data center to compile and evaluate the data for the above reactions as soon as possible.

- All (reference) standard sources of radioactivities used for the calibration of detectors should be traceable to the IAEA or other reference laboratories recognized by the IAEA.

- The consultants were in unanimous agreement that all experimental data should be reported to journals and nuclear data banks in the form of cross-sections. A period of tolerance for the barn (for example until 1990) would be acceptable before the complete imposition of SI units.
No! The barn should be recognized as an SI unit! (HDL)

- The topic of how to report practical thick target yields was more controversial. This has remained so since the publication and discussion summary of Svoboda and Silvester at the Oxford meeting in 1969(1). For the moment the selection of the practical unit (in terms of the way the data are to be used) should remain at the discretion of the author, although there was a strong focus toward the adoption of SI units. Reporting of the integral yield or production yield curve at saturation vs energy was considered useful to the MRI radiochemists.

- The working group asks the IAEA to communicate to experimentalists and editors of journals that publications should provide relevant experimental details about target preparation, including details of target composition, chemical form, isotopic abundance, preparation and containment and the estimation of the number of target atoms. Information about recoil range distribution, and beam power density effects on targets should also be described.

- Cross-section data for several medical radioisotopes are well documented. However, with increasing energy the data needs also increase. Many existing data are adequate, but in some cases more experimental measurements are needed. Due to the fact that each

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(1) Amphlett, C.B. (ed.), The Uses of Cyclotrons in Chemistry, Metallurgy & Biology, Proc. Conf. held at St. Catherine's College, Oxford, 22-23 Sept. 1969, Butterworth, London, 1970. Also K. Svoboda and D.J. Silvester presented their paper "Quantities and Unit Used in the Production of Radionuclides by Charged Particle Bombardment" to Int. J. Appl. Radiat. Isotopes 22 (1971) 269.

institution is in a different situation as regards the maximum particle energy available at a given accelerator, the types of the particles accelerated, the chemical form and enrichment of the target, only individual data needs could be identified. It was recognized that there are deficiencies in excitation functions for a number of medical radioisotopes. It is believed that work is in progress in some laboratories on some of these reactions.

$^{11}B(p,n)^{11}C$
$^{13}C(p,n)^{13}N$
$^{38}Ar(p,n)^{38}K$
$^{40}Ar(p,3n)^{38}K$
$^{77}Se(p,n)^{77}Br$
$^{82}Kr(p,2n)^{81m+g}Rb$
$^{nat}Rb(p,xn)^{82}Sr$
$^{92}Mo(p,n)^{92}Tc$
$^{100}Mo(p,2n)^{99m}Tc$
$^{112}Cd(p,2n)^{111}In$
$^{123}Te(p,n)^{123}I$
$^{124}Xe(p,2p)^{123}I$
$^{124}Xe(p,pn)^{123}Xe$
$^{124}Xe(p,2n)^{123}Cs$
$^{74}Se(d,n)^{75}Br$
$^{82,83,84}Kr(d,xn)^{81}Rb$
$^{nat}Rb(d,xn)^{82}Sr$
$^{82,83,84}Kr(^{3}He,xn)^{82}Sr$
$^{82,83,84}Kr(^{4}He,xn)^{82}Sr$

- The consultants concluded that it would be useful if the IAEA in collaboration with other nuclear data centers would look criticially at the experimental information available on cross-sections and production yields for the more commonly used medical radioisotopes. These include ^{11}C, ^{13}N, ^{15}O, ^{18}F, ^{67}Ga, ^{111}In, ^{123}I, ^{201}Tl.

- The decay data for medical radioisotopes are in most cases well known and documented. However, during the meeting several radioisotopes were identified for which minor revision or verification of the available decay data would be desirable. These are ^{55}Co (half-life uncertainty), ^{52m}Mn, ^{77}Br, ^{62}Zn, ^{63}Zn, ^{66}Ga, ^{75}Kr, ^{77}Kr, ^{81m}Rb, ^{123}Xe, $^{195m+g}Hg$ - ^{195m}Au (branching ratios, γ-ray abundances, etc.). The IAEA is asked to bring these deficiencies to the attention of the relevant bodies.

- The working group encouraged the development of a compilation of nuclear data for medical radioisotopes produced by accelerators such as that independently initiated in 1981 by Qaim[2] and recently updated by NDS[3]. The working group proposed that the compilation should include all medical radioisotopes of current interest, and exhaustive references of all reported cross-section and production yield related details. A suggestion is to include threshold energies and thick target yields and to add the following radionuclides to the list prepared by NDS[3].

  ^{47}Ca, ^{67}Cu, ^{66}Ga, ^{75}Se, ^{77}Kr, ^{79}Kr, ^{89}Zr, ^{95m}Tc, ^{96}Tc, ^{100}Pd, ^{101m}Rh, $^{107}Cd_T$ ^{107m}Ag, ^{124}I, ^{157}Dy, ^{117m}Sn, ^{127}Xe, ^{169}Yb, ^{186}Re, $^{211}Rn_T$ ^{211}At, ^{205}Bi, ^{206}Bi.

- The working group strongly supported the continuation of various IAEA activities in medical radioisotope production. The Agency should look at the matter at some intervals at the Consultants' level. Further more it was felt that a joint seminar dealing with nuclear data, technological and radiochemical aspects of accelerator radioisotope production (possibly in collaboration with the nuclear data, chemistry and physics sections of the Agency) be held around 1989.

- The question of enriched stable isotopes for medical radioisotope production was addressed by the Agency's Consultants' Meeting held in Turku in 1985[4]. This working group agrees that there are presently no serious difficulties in obtaining the commonly used highly enriched target materials for the production of ^{11}C, ^{13}N, ^{15}O, ^{18}F and ^{201}Tl. However, certain highly enriched stable isotopes required for medical radioisotope research and development are not readily available, e.g. ^{122}Te, ^{123}Te, ^{124}Xe.

- The working group recognized the predictive value of computer codes based upon nuclear models. It was, however, strongly emphasized that the codes should be user oriented, and that the Agency should encourage access to these codes.

- A wide distribution of this report by the IAEA is strongly recommended, (a detailed list of recipients will be augmented by the working group).

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(2) S.M. Qaim, Radiochimica Acta 30 (1982) 147-162

(3) D. Gandarias-Cruz and K. Okamoto, presented at this meeting "Nuclear Data for Medical Radioisotopes Produced by Accelerators - Status and Compilation".

(4) Summary Report of the IAEA Consultants' Meeting on "Cyclotron Production of Radionuclides with Enriched Targets", Turku, Finland, 22-25 July 1985 (edited by H. Vera-Ruiz).

Conclusions

The Consultants' working group on experimental data recommended the following actions on the part of the Agency. (1) The compilation and evaluation of data for nuclear reactions used to monitor accelerator beam energy and intensity as soon as possible, (2) the evaluation of cross sections and production yields for the commonly used accelerator produced medical radioisotopes, (3) the transmission of deficiencies in certain excitation functions and nuclear data to the relevant bodies, (4) the compilation of nuclear data for all accelerator produced medical radioisotopes, (5) the holding of a seminar on nuclear data, technological and radiochemical aspects of medical radioisotope production around 1989, and (7) the dissemination of the results of the discussion on the standardization of the cross-section measurements to experimentalists, journal editors, and libraries.

Working Group II:

CALCULATION AND COMPUTER FILE OF EXCITATION FUNCTIONS

M. Blann (Chairman), A. Pavlik, K. Hata, M.C. Lagunas-Solar
S.M. Qaim, K. Sueki

SUMMARY

We have heard a well balanced and versatile range of presentations spanning the range of accelerator production of radioisotopes for medical applications. Here we wish to say a few words about the presentations dealing with nuclear model codes as a tool for use in radioisotope production.

There was unanimous agreement that the final answer in obtaining necessary reaction cross sections, if feasible, should always be experimental. Yet, we also saw in many of the research end of the field the wish to understand reaction cross sections in terms of nuclear reaction models, and perhaps eventually to be able to use these models as a guide in selecting the best experimental conditions for producing a given radioisotope without having to measure all the possible reactions.

Along these lines we heard preliminary efforts by Lagunas-Solar in interpeting his yields of F-18 with ALICE code. Lu Hanlin and his co-workers at Beijing have the most impressive agreement with the experimental excitation functions around A=87 region using a code they wrote based on the hybrid precompound and Weisskopf-Ewing evaporation models. We hope to get more information on this fine code.

The RIKEN workers, given in the papers by Tendow, Kitao and Sueki, have also illustrated the comparisons of calculated excitation functions with several data sets. The question was raised as to whether the calculations may sometimes not be of a help to select the better data set when there are large disagreements between several sets.

A major step beyond the usual statistical/precompound codes was presented by Hata of JAERI in the OSCAR code. This is really a code system designed to give the thick target yield information necessary to most medical radioisotope production problems. It includes the use of experimental thin target yields as input when available, supplemented by results from systematics from the ALICE nuclear model code where experimental results are not available. A contribution from Masumoto suggests algorithms for computing yields for photonuclear reactions.

There were two additional papers stressing the use of nuclear theory. Pavlik gave a summary of physics of several of the popularly used codes. He went on to illustrate the excellent results which could be obtained by careful applications of STAPRE (Hauser-Feshbach plus exciton model) code of Uhl. The latter is internationally recognized as the best code in the class. Pavlik showed predictive power to within 30% of experimental yield. We hasten to point out that experimental yields may often have systematic errors approaching this limit.

Blann presented comparisons of the results of ALICE code with a broad range of nuclear reactions, mostly proton and light ion induced, at energies up to 200 MeV. The calculated results shown were generally done by the experimental groups who measured the relevant excitation functions. The philosophy of ALICE code (Weiskopf-Ewing plus hybrid model) differs from that of STAPRE, in that it is not intended to give the best possible calculation, but rather it stresses ease of use by non-experts, and short computer time requirements. There is agreement between the Vienna and Livermore code groups that STAPRE is the code of choice when the highest possible accuracy (30% region) is desired, and ALICE is useful when a factor of two or better is adequate, and when the user wishes to keep the investment in time to run the codes at a minimum.

Some conference participants felt that the capability of running model codes in their laboratories would be valuable. With many codes in existence, the IAEA could help in the selection of process by summarizing important facts on the availability and capability of each code. Working Group II has prepared specific recommendations in this regard.

RECOMMENDATIONS

The working group feels that computational capability of excitation functions is a valuable tool to help guide ultimate experimental programs in radioisotope production for medical applications.

The IAEA or a related organization could provide a very valuable service to this community in several ways, the goal being to aid in the selection and determination of availability of these codes.

In order to evaluate the suitability of the many codes in existence, and to efficiently select the code or codes best fitting each laboratory's needs, a compilation of answers to a few questions would provide a valuable service. An initial set of questions is as follows:

(1) What codes are presently available which are suitable for general use for calculating excitation functions, and to whom does one write to receive a copy of these codes? Is there any cost? Are the codes written entirely in for example, a standard FORTRAN language?

(2) For each code available, is there a manual or some documentation explaining how to run the code?

(3) Are sample input and output available for checking the operation of the code on the end user's computer? What are the options on input and output?

(4) What physics is in the code - e.g. Wiesskopf or Hauser Feshbach, type of precompound decay; are γ-ray cascades treated?

(5) What is the maximum excitation energy which the distributed version of the code will accommodate?

(6) What is the range of ΔA and ΔZ of product nuclei (i.e. how many neutrons and protons may be emitted in a single calculation?

(7) Are all reaction paths treated in a 'single pass', or are multiple passes required?

(8) What projectiles may be accommodated in the entrance channel and in the exit channel?

(9) Are discrete levels allowed in the input? Are they required?

(10) How are level densities handled?

(11) Give a sample input for some test problem.

(12) What is the computer core requirement and running time on the author's computer? Is the code known to run on any smaller computer?

(13) Is the code written to run in batch mode or in interactive mode?

A second valuable result to present would be an intercomparison of several of these codes in calculating several excitation functions. Recommended are the $^{127}I(p,xn)$ and $^{75}As(p,xn)$ excitation functions at incident proton energies up to 70 MeV. It would be nice to have the sample input for each code required to run these tests, and the computer type and CPU time used. These calculations should be performed for incident protons in 2 MeV energy increments. Results of all calculated excitation functions should be shown graphically, together with the experimental results on the same graphs.

A resonably complete list of codes/authors/users was sent from Livermore (M. Blann) as follows:

STAPRE (IRK)	S. Wilboolsak, B. Strohmaier, M. Uhl, Vienna
STAPRE (LLL-1)	D.G. Gardner, M.A. Gardner, LLNL, Livermore
GNASH (LAS)	P.G. Young, LANL (N.B. - there are several) GNASH- HF, WE, Los Alamos
EMPIRE (IBJ)	M. Herman, ENEA, Bologna; A. Marcinkowski, Warsaw University, Warsaw
PERINNI (ECN-1)	H. Gruppelaar, H.A.J. van der Kamp, Petten
HAUSER-V (TRM-1)	S.B. Garg, A. Sinha, BARC, Trombay Bombay
TNG (ORL)	C.Y. Fu, ORNL, Oakridge
PRANG (ECN-2)	H. Gruppelaar, H.A.J. van der Kamp, Petten No γ-ray competition included (future option: see GRYPHON code, Ref. H. Gruppelaar, I.M. Akkerman, Int. Conf. on Nuclear data for Basic and Applied Science, Santa Fé, USA, May 1985 and Rep. ECN 164)
PEOGM (SLO)	E. Bevtak, Bratislava
GNASH (JAE)	K. Shibata, E. Arthur, P.G. Young, Tokai, LANL
PREM(TOH)	G. Keeni, S. Yoshida, Sendai
PREANGL1 (TRM-2)	S.B. Garg, A. Sinha, BARC, Trombay Bombay
AMAPRE (TUD)	H. Kalka, D. Hermsdorf, D. Seeliger, Dresden
SECDIST (KFK)	I. Broeders, U. Fischer, H. Jahn, E. Wiegner, Karlsruhe

PRECO-D2 (TNL)　　C. Kalbach, U. North Carolina
PENELOPE (IDA)　　G. Reffo, ENEA, Bologna
ALLICE (LLL-2,-3)　　M. Blann, LLNL, Livermore
HELGA (MPI)　　H. Klapdor, Heidelberg

These exercises are intended to inform the medical radioisotope producer community as to which codes are available to them, and which are compatible with their specific needs and computational resources. The second part is intended to show the relative merit of the output of these codes, which is an important criterion in deciding which code or codes to implement.

PRECO-D2 (INL) C. Kalbach, ... Basic Calculus
PENELOPE (IDA) **Working Group III: DATA COMPILATION**
ALLICE (LLL-2,-3) M. Blann, LLNL, Livermore
HELGA (MPI) **(including Proposal for an IAEA Handbook on "Data for Medical Radioisotope Production"**

These exercises are intended to inform the medical radioisotope producer which are compatible with the second part of these codes, which ... to implement.

D. Berényi (Chairman), G.J. Beyer, M. Blann, M. Bonardi, A. Hashizume, M.C. Lagunas-Solar, R.M. Lambrecht, Lu Hanlin, T. Nozaki, A. Pavlik, S.M. Qaim, S.L. Waters, K. Hata, J. Heinzl, S. Igarasi, K. Kitao, H. Morinaga, Y. Ohkubo, K. Sueki, Y. Tendow, H. Umezawa

The Working Group III held its session in the form of a plenary meeting and discussed the issue of the compilation of the most important nuclear data for the medical radioisotope production by accelerators and that of a proposed IAEA Handbook on Nuclear Data for Medical Radioisotope Production.

After some discussion it was concluded that it is not timely and suitable to publish a compilation and a handbook separately.

It is suggested to make one publication with the title "Handbook on nuclear data for medical radioisotope production". The contents could be as follows: In a short introduction the most important nuclear concepts on radioisotope production by accelerators should be clarified (in a "definition-like" way). Then comes a section with evaluated cross section data for the production of some of the most important medical radioisotopes (e.g. the lightest positron emitting isotopes for PET studies, namely ^{11}C, ^{13}N, ^{15}O, and ^{18}F and other isotopes such as ^{67}Ga, ^{111}In, ^{123}I, ^{201}Tl) as well as those for monitor reactions. In this section the rules for standardization of the pertinent data should also be included (see the Report of WGI) which would be guide-line for the evaluators but - at the same time - it might promote to measure and publish the data concerned by the authors in the future in a more unified and more complete way. It is also expected that the evaluation work on the data for production of most important medical radioisotopes will show the shortages and inconsistencies in the pertaining data and so it can initiate further experimental studies in some cases.

Finally, an important part of the handbook would be a rather detailed and somewhat critical tabulation (a compilation) of the data for the production of medical radioisotopes (see some details in the Report of WGI).