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First Research Co-ordination Meeting on

DEVELOPMENT OF REFERENCE CHARGED PARTICLE CROSS SECTION DATA BASE FOR MEDICAL RADIOISOTOPE PRODUCTION

Vienna, 15 - 17 November 1995

SUMMARY REPORT

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Abstract

The present report contains the summary of the First Research Co-ordination Meeting on "Development of Reference Charged Particle Cross Section Data Base for Medical Radioisotope Production", held at the IAEA Headquarters, Vienna, from 15 to 17 November 1995. The project focuses on monitor reactions and production reactions for gamma emitters and positron emitters induced with light charged particles of incident energies up to about 100 MeV. Summarized are technical discussions and the resulting work plan of the Coordinated Research Programme, including actions and deadlines. Attached are an information sheet on the project, the agenda and a list of participants of the meeting. Also attached is brief information on the adjacent Consultants' Meeting on "Automated Synthesis Systems for the Cyclotron Production of ^{18}F and ^{123}I and their Labeled Radiopharmaceuticals".

March 1996

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I. Summary of the Meeting

The First Research Co-ordination Meeting on "Development of Reference Charged Particle Cross Section Data Base for Medical Radioisotope Production" was held at the IAEA Headquarters, Vienna, Austria, from 15 to 17 November 1995. The related Coordinated Research Programme (CRP) focuses on beam monitor reactions and production reactions for positron emitters and gamma emitters induced by light charged particles (protons through alpha-particles) with incident energies up to about 100 MeV. The main objective of the meeting was to work out the detailed scope and a related work plan of the CRP.

The meeting was attended by 6 out of 7 chief scientific investigators of the CRP and by 4 observers. Prof. S.M. Qaim (Forschungszentrum, Institut für Nuklearchemie, Jülich, Germany) served as Chairman of the meeting. The agenda and list of participants of the meeting are attached *(Appendix 1)*.

The conclusions and recommendations of the Research Coordination Meeting including a detailed work plan, are given below. Attached is the Information Sheet on the CRP *(Appendix 2)*.

The venue and dates of the 2nd Research Co-ordination Meeting were discussed. Dr. S.J. Mills (National Accelerator Centre, Faure, South Africa) has kindly offered his services to host the meeting. March/April 1997 was found to be the best dates for the 2nd Research Co-ordination Meeting.

The present meeting was organized adjacent to the IAEA Consultants' Meeting on "Automated Synthesis Systems for the Cyclotron Production of ^{18}F and ^{123}I and their Labelled Radiopharmaceuticals", Vienna, 13-16 November 1995 *(see Appendix 3 for Summary and Recommendations)*.

II. Opening Session

The first session was held (on the forenoon of 15 November 1995) jointly with the Consultants' Meeting on "Automated Synthesis Systems for the Cyclotron Production of ^{18}F and ^{123}I and their Labelled Radiopharmaceuticals". After the opening remarks by the secretaries of the two Meetings (Pavel Obložinský - Nuclear Data Section and Hernan Vera-Ruiz - Industrial Applications and Chemistry Section), Syed M. Qaim (KFA Jülich, Germany) was elected as Chairman and the suggested Agenda (see *Appendix 1*) was adopted without any modification. Thereafter three review lectures, expected to be of common interest to participants of both the groups, were presented.

In the first lecture **Syed M. Qaim** (KFA Jülich, Germany) gave an overview of the general status of nuclear data relevant to cyclotron production of medically important radioisotopes, as well as an in-depth review of the data for the production of β^+ emitters and therapy related radionuclides. In general, for the commonly used β^+ emitters the available cross section information is relatively extensive. However, many of the data are discrepant and an evaluation of the known data is needed. A considerable amount of nuclear data research is needed when suitable production processes for potentially useful β^+ emitters are being investigated. The production of therapeutic radioisotopes may also involve some nuclear data work, especially if the radionuclide under consideration is to be produced at a cyclotron. The field of endoradiotherapy is a developing field and it demands rather strong effort on the part of a chemist. The nuclear data effort involved is, however, rather small.

In the second lecture **Richard M. Lambrecht** (University of Tübingen, Germany) discussed the nuclear data relevant to the production of radioisotopes used in single photon emission tomography (SPET). Special consideration was given to the most commonly used radioisotopes ^{123}I and ^{99m}Tc. The data for ^{123}I are extensive but there are some discrepancies in the case of the commonly used ^{124}Xe(p,x)^{123}I process. The case of ^{99m}Tc was discussed in more detail.

The problem of the future availability of nuclear reactors suitable for ^{99}Mo production has prompted a renewed interest on alternative production methods. Three possible cyclotron production routes (one for ^{99}Mo and two for ^{99m}Tc) were discussed:

- the low energy production of ^{99m}Tc through the ^{98}Mo(p,γ)^{99m}Tc reaction (cross section too low),

- the medium energy production of ^{99m}Tc via the ^{100}Mo(p,2n)^{99m}Tc reaction, and

- the high energy production of ^{99}Mo via the ^{100}Mo(p,pn)^{99}Mo reaction or via alternative reactions.

The technical feasibility of these production methods is still being evaluated. Among others, there are issues regarding radionuclides impurities, specific activities, targetry, distribution logistics, licensing process and economics.

In the third lecture **Pascal Cohilis** (IBA, Louvain La Neuve, Belgium) described some recent developments in the manufacturing of high intensity cyclotrons which find applications in the production of radioisotopes for diagnostic nuclear medicine or for radiotherapy. The first cyclotrons for radioisotope production were classical, with the extracted beam intensity limited to 100 μA or less. Internal targets were used when higher currents were needed. In 1987, IBA introduced the CYCLONE 30, featuring an innovative magnet design and an external ion source. Currents for such machines were increased with time to more than 1 mA extracted, with 2 or 3 mA in view. For high intensity applications without extraction, positive ion acceleration still presents significant advantages, as illustrated by the CYCLONE 18$^+$, operating daily at 2 mA continuously on an internal target. Space charge calculations indicate that the intensity limit due to the axial component of the space charge forces is probably around 10 mA average beam current. This applies not only to positive ion cyclotrons using an internal target but also to negative ion cyclotrons where the extraction is made by charge exchange.

The last part of the Session was devoted to a discussion of the status of medically oriented cyclotrons. The initiative of Hernan Vera-Ruiz (IAEA, Industrial Applications and Chemistry Section) to compile a Worldwide Directory of Cyclotrons used for radionuclide production was appreciated. In recent years more information has been made available on some older cyclotrons in Russia and in the former Soviet Republics. Furthermore, new cyclotron centres have been established in several developing countries like Argentina, Indonesia, Iran etc. Thus a compilation of the Directory appears very timely. It was suggested that the Questionnaire prepared by the Consultants' Meeting should include a further question related to the measurement and use of nuclear data. It was felt that the joint session proved to be very useful to both the groups.

III. Laboratory Reports

Reports were presented from each laboratory participating in the CRP.

VUB Cyclotron Laboratory, Brussels, Belgium
(Alex Hermanne)

At the VUB Cyclotron laboratory a multiparticle (p, d, α, ^{3}He) CGR-560 accelerator has been in operation for 10 years. The maximum proton energy is about 43 MeV (depending on deflector settings) and up to 120 μA of beam current could be obtained on external targets.

Starting in 1987, on demand of radiopharmaceutical companies, a systematic study of optimized production routes (irradiation procedure, target preparation, automated chemistry which is now commercially available through IBA) was initiated for $^{201}T\ell$, ^{67}Ga and ^{111}In. For those nuclides cross sections were analyzed and measured for several reactions leading to the final products ($^{203}T\ell(p,3n)^{201}Pb$; $^{68}Zn(p,2n)^{67}Ga$ and $^{112}Cd(p,2n)^{111}In$) as well as most of the contaminating reactions on the natural target materials in the energy range from threshold to 42 MeV.

Cross sections for production of Br radioisotopes through α-reactions on As and of Co radioisotopes through p-reactions on Ni and Co-foils were also determined. An effort to reduce the uncertainty on the beam energy was done by using a combination of 3 techniques: charged particle TOF, stopping power analysis, and activation analysis.

In recent work on recoil effects and on a systematic study of monitor reactions a reassessment of the status of several processes was done: $^{nat}Ni(p,x)^{57}Ni$, ^{56}Ni, ^{56}Co, ^{55}Co with good agreement with literature values; $^{nat}Cu(p,x)^{62}Zn$, ^{65}Zn with reasonable agreement, and $^{nat}A\ell(p,x)^{22}Na$ with good agreement. For α-particle beams $^{nat}Cu(\alpha,n)^{66}Ga$, ^{67}Ga, ^{65}Zn were studied with varying results while $^{nat}Ti(\alpha,n)^{51}Cr$ data are coincident with published values. Values for $^{27}A\ell(d,x)^{24}Na$ and $^{nat}V(d,x)^{51}Cr$ are also available.

China Nuclear Data Center, Beijing, P.R. China
(Zhuang Youxiang)

The status of work on Charged Particle Nuclear Data (CPND) compilation and evaluation at China Institute of Atomic Energy (CIAE) was reported.

CNDC-Charged Particle Group has compiled a large number of experimental data measured in China and submitted them to the IAEA Nuclear Data Section.

As far as evaluation is concerned, neutron data are more numerous and have higher accuracy than charged particle ones. Therefore, research on evaluation methodology for charged particle nuclear data is especially important. A close combination of evaluation with measurements and theoretical calculations is essential.

Through many years CNDC-Charged Particle Group at CIAE has developed a series of calculational code systems for charged particle nuclear data, such as CMUP2, CFUP1, CUNF, SPEC, DDCS, evaluated some activation cross sections as well as completed sets of CPND, and summarized "Techniques Used for Charged Particle Nuclear Data Evaluations at CNDC" which is published in the Proceedings of the International Symposium on Nuclear Data Evaluation Methodology, BNL, USA, 1992, p. 575-584.

A new 30 MeV proton cyclotron was established at CIAE in 1994, and some radioisotopes for medical application ($^{67,68}Ga$, ^{68}Ge, ^{109}Cd, ^{111}In, $^{201}T\ell$) have been produced.

Forschungszentrum (KFA) Jülich, Institut für Nuklearchemie, Jülich, Germany
(Syed M. Qaim)

There are three cyclotrons at Jülich which are exclusively or partly used for medical radioisotope production. The Baby Cyclotron BC1710 is a dedicated machine for radiopharmaceutical production for Positron Emission Tomography (PET). The Compact Cyclotron CV28 is a multipurpose multiparticle variable energy machine. About 50% of the irradiation time stands at the disposal of the Institute of Nuclear Chemistry. The radioisotope production group has also access to the Injector Cyclotron of the Cooler Synchrotron (COSY) located in the Institute of Nuclear Physics.

An extensive research and development programme related to medically important cyclotron radioisotopes has been underway for the last 20 years. It involves work in all branches of radiopharmaceutical development, e.g. nuclear data measurement, targetry, radiochemical separation, labeling of biomolecules with radioisotopes, biological evaluation of tracer (animal experiments), and finally applications in humans. As far as nuclear data activities are concerned, the emphasis has been on investigation of production routes for radioisotopes used in emission tomography (i.e. Positron Emission Tomography - PET and Single Positron Emission Tomography -SPET). In recent years increased attention has also been devoted to therapy related radioisotopes.

Institute of Nuclear Research of the Hungarian Academy of Sciences, Cyclotron Application Department, Debrecen, Hungary
(Ferenc Tárkányi)

The Cyclotron Laboratory was established in 1985. The main task of the Department is to use the cyclotron in different fields, e.g. medicine, agriculture and industry. Some of them include:

- production of radioisotopes for medical and biological investigations (^{11}C, ^{15}O, ^{18}F, ^{67}Ga, ^{123}I)
- activation analysis and wear measurement using thin layer activation technique (Fe, Ni, Ti, Mo, etc.)
- effects of fast neutrons on biological samples and electronic components
- charged particle nuclear data for applications
- targetry and investigation of basic processes in targets

The Department uses the technical base of the Institute (MGC-20E cyclotron, PET, nuclear spectrometers) and the scientific cooperation with other institutes participating in this work (KFA Jülich, Abo Academy, VUB Brussels). The staff includes 7 physicists, 4 chemists an 2 technicians.

The main activities in nuclear data are:

a) Measurements of excitation functions of reactions used for monitoring the beam performances and for isotope production purposes

b) Compilation and evaluation of nuclear data in EXFOR and other formats.

Institute of Physics and Power Engineering, Department of Nuclear Physics, Theoretical Division, Obninsk, Kaluga Region, Russia
(Yuri Shubin)

The Theoretical Division is engaged in evaluation of different types of nuclear data. A small group deals with the evaluation of production data relevant to medically important radioisotopes. In general, ALICE code with several new modifications is used for evaluation purposes.

A report on Cyclotron Co. Ltd. in Obninsk was also presented. The cyclotron was put into operation in 1963, mainly for radionuclides production. The improvements in accelerator, targets and radionuclide preparation techniques turned the Cyclotron company into the main supplier of the cyclotron produced radionuclides and radiation sources in Russia. The cyclotron accelerates protons and deuterons up to 21 MeV with beam currents on target up to 1 mA.

Main directions of the enterprise are:

- development of methods and technologies of cyclotron radioisotope production
- manufacture specified radioisotopes

Isotopes are delivered as solutions. Main products are: ^{57}Co, ^{67}Ga,^{68}Ge, ^{85}Sr, ^{109}Cd, ^{111}In, ^{181}W, ^{207}Bi, intended for use in nuclear medicine, industry, scientific research and other special purposes.

Investigations of production techniques for new radionuclides are being carried out continuously. It is planned to start the production of the radionuclide ^{201}Tl in the beginning and ^{123}I in the middle of 1996. Production technique for ^{67}Cu of high radionuclide purity is also under development.

National Accelerator Centre (NAC), Faure, South Africa
(Stephen Mills)

The NAC is a multidisciplinary facility, operated by the South African Foundation for Research Development as a national, user-based research centre for the country. It provides accelerator and auxiliary facilities for basic and applied research in the physical and bio-medical sciences, neutron and proton radiotherapy for the treatment of cancer and the production of a large variety of radioisotopes. The accelerators operated by NAC are a variable-energy K=200 separated-sector cyclotron with two K=8 solid-pole injector cyclotrons, one of which provides high-intensity light-ion beams and the other heavy-ion beams and polarized light ions, as well as a 6 MV Van de Graaff accelerator.

The NAC cyclotron is almost without doubt the best utilized research facility in South Africa, being used 24 hours per day, seven days per week and averaging about 18 hours of useful beam time per day over the whole calendar year. This beam time is at present about equally divided between physics research, production of radioisotopes and therapy and calibrations. NAC's high-energy neutron therapy facility is the only such facility in Africa as well as the Southern Hemisphere and is considered one of the best in the world, based on its optimum beam energy (66 MeV), isocentric treatment head and adequate infrastructure, including a 30-bed on-site hospital. More than 780 patients have now been treated, with some spectacular successes, e.g. for salivary gland and advanced breast cancers. South Africa is also one of only 7 countries in the world where high-energy proton therapy can be performed, and more than 100 patients have already been treated since September 1993 on NAC's horizontal-beam proton radioneurosurgical facility. NAC is also at present the only centre in the world where high-energy protons as well as high-energy neutrons are available for therapy on the same site.

The radioisotopes which are produced at NAC satisfy the entire country's demand for certain important medical radioisotopes like ^{18}F, ^{67}Ga, $^{81}Rb/^{81m}Kr$, ^{111}In, ^{123}I and its labeled compounds and ^{201}Tl. These are supplied regularly to more than thirty provincial hospitals, private nuclear medicine practices and research institutions and are used in nearly 10,000 patients each year. Some long-lived non-medical radioisotopes, such as ^{22}Na and ^{55}Fe, are also produced, mostly for export. NAC will also be the only possible producer of positron-emitting radioisotopes when South Africa in future acquires a PET camera.

The radioisotope programme at NAC is based on a 66 MeV proton beam, which is shared with neutron therapy during a large part of the week. Special attention has therefore been given to the determination of excitation functions for the production of radioisotopes in the higher energy region. More than 160 excitation functions in total have thus far been measured up to either 100 or 200 MeV for proton bombardment of, inter alia, natural Aℓ, Mn, Ni, Cd, In, Zn, Ge, Kr, Cu and Xe. The analysis of data obtained for Pr+p up to 100 MeV is at present in progress, while measurements on Ag+p are planned for the near future. In most cases the measured excitation functions have been compared with theoretical calculations performed by means of ALICE/85/300. This programme can also rely on the services of the NAC Radioactivity Standards Laboratory, which is a member laboratory of both the International Bureau of Weights and Measures (BIPM) in Paris, France, and the International Committee for Radionuclide Metrology (ICRM), and regularly participates in international intercomparisons of radioactivity measurements with excellent results.

IV. Technical Discussions

1. Monitor Reactions

Discussion Leader: *Ferenc Tárkányi*

Monitor reactions play an important role in the determination of various parameters (like energy, intensity, etc.) of the charged particle beam. The use of monitor reactions constitutes the simplest method and yet it assures the necessary precision needed in different applications. The other methods developed for measuring currents of charged particle beams (Faraday-cup, beam current transformers, calorimetric methods) are more difficult to apply. Similarly the procedures used for determining the energy of the bombarding beam (TOF, Rutherford back-scattering, magnetic spectrometers, range-energy functions, calibrated analysing magnets, spatial separation between neighbouring bunches, etc.) are technically more sophisticated, and need more expertise in applications.

There are different methods for use of monitor reactions in the determination of beam parameters (absolute and relative methods, one or two parameter method, etc.). All those methods demand as a prerequisite well known and reliable excitation functions of some monitor reactions.

There are several other criteria regarding the monitor targets and monitor reactions. These include the availability, the physical and chemical characteristics of the target material, the decay characteristics of the produced nuclides, the slope of the excitation function, and the undesired interfering reactions.

Taking into account all these requirements, the following target materials could be considered as good candidates: aluminium, nickel, titanium, copper and iron. It is also favourable to use the same material (if possible) for all the particles used in isotope production (p, d, ^{3}He, α). A critical compilation and evaluation of the existing data needs to be carried out.

Considering all the above-mentioned parameters and the available cross-section data it is recommended that the following monitor reactions be given strong consideration. A critical compilation and evaluation of the existing data needs to be carried out.

Monitor reactions for protons

$^{nat}A\ell(p,x)^{22}Na$
$^{nat}Ti(p,x)^{48}V$
$^{nat}Ni(p,x)^{57}Ni$
$^{nat}Cu(p,x)^{56}Co$
$^{nat}Cu(p,x)^{62}Zn$
$^{nat}Cu(p,x)^{63}Zn$
$^{nat}Cu(p,x)^{65}Zn$

Monitor reactions for deuterons

$^{nat}A\ell(d,x)^{22}Na$
$^{nat}Ti(d,x)^{48}V$
$^{nat}Fe(d,x)^{56}Co$
$^{nat}Ni(d,x)^{61}Cu$

Monitor reactions for ^{3}He-particles

$^{nat}A\ell(^{3}He,x)^{22}Na$
$^{nat}Ti(^{3}He,x)^{48}V$
$^{nat}Cu(^{3}He,x)^{66,67}Ga$, ^{65}Zn

Monitor reactions for α-particles

$^{nat}A\ell(\alpha,x)^{24}Na$
$^{nat}Ti(\alpha,x)^{51}Cr$
$^{nat}Cu(\alpha,x)^{66,67}Ga$, ^{65}Zn

The desired accuracies of the evaluated data are not very high. Recommended values with errors of 6 to 8% would be acceptable.

2. Positron Emitters

Discussion Leader: *Syed M. Qaim*

In recent years Positron Emission Tomography (PET) has been gaining considerable significance in diagnostic nuclear medicine, especially for investigating physiological functions in vivo. Since it constitutes a fast and quantitative method for studies on humans, its applications are increasing. A large number of PET Centres have been established in more advanced countries and the technique is being recognised as an important diagnostic tool.

The success of the technique is dependent on the availability of suitable β^{+} emitting radiopharmaceuticals. Thus a considerable amount of research work is underway to develop a variety of radiopharmaceuticals. The major β^{+} emitting radioisotopes used in the labelling of biomolecules are $^{11}C(t_{1/2}=20min)$ and $^{13}N(t_{1/2}=10min)$, $^{15}O(t_{1/2}=2min)$ and $^{18}F(t_{1/2}=110min)$. Due to the very short half-life of the first three radioisotopes, the production and application of the respective radiopharmaceuticals has to be done on site. In the case of ^{18}F the transportation of the radiopharmaceutical is economically possible to nearby centers.

Besides the four major short-lived organic β^+ emitters, there are a few generator systems which constitute some very convenient sources of β^+ emitting daughters. These are $^{62}Zn(t_{1/2}=9.3h)$ / $^{62}Cu(t_{1/2}=10min)$, $^{68}Ge(t_{1/2}=288d)$ / $^{68}Ga(t_{1/2}=68min)$ and $^{82}Sr(t_{1/2}=25.5d)$ / $^{82}Rb(t_{1/2}=1.2min)$. Furthermore, there are a large number of other potentially important β^+ emitting radioisotopes.

The CRP gave a brief consideration to the various β^+ emitters, but concentrated especially on the four short-lived organic β^+ emitting radioisotopes as well as the three generator radionuclides. A brief résumé of the discussion is given below.

$^{11}C(t_{1/2}=20$ min)

Several routes are available for the production of this radioisotope. The most common method, however, is the $^{14}N(p,\alpha)$-process. Considerable amount of cross section information is available. However, no evaluation has been done so far. The task is rather difficult since a large number of resonances have been observed.

$^{13}N(t_{1/2}=10$ min)

For this radioisotope as well several nuclear reactions have been suggested. The most common route, however, is the $^{16}O(p,\alpha)$-process. Although several measurements exist, no evaluation has been attempted.

$^{15}O(t_{1/2}=2$ min)

The most common method for the production of this radioisotope is the $^{14}N(d,n)$-reaction. The case is similar to ^{13}N.

$^{18}F(t_{1/2}=110$ min)

This radioisotope is commonly produced via the $^{20}Ne(d,\alpha)^{18}F$ and $^{18}O(p,n)^{18}F$ reactions. Whereas the former reaction yields $[^{18}F]F_2$ the latter gives $^{18}F^-$. Both the chemical forms are suitable starting materials for subsequent labelling work. Whereas several measurements exist for the $^{20}Ne(d,\alpha)$-reaction, in the case of $^{18}O(p,n)^{18}F$ process only one measurement has been done. It is considered worthwhile to do an evaluation of the $^{20}Ne(d,\alpha)$ data but in the case of the $^{18}O(p,n)^{18}F$ reaction it may be very useful to measure the averaged cross section over small energy intervals.

$^{62}Zn(t_{1/2}=9.3$ h)

This parent generator radioisotope (mentioned above) is being considered in the context of a monitor reaction product.

^{68}Ge($t_{½}$=288d) and ^{82}Sr($t_{½}$=25.5d)

For the production of ^{68}Ge, in addition to spallation, the ^{69}Ga(p,2n) and ^{nat}Ga(p,x) processes have been suggested. Similarly for ^{82}Sr the ^{85}Rb(p,4n) and ^{nat}Rb(p,x) reactions have been used. Several cross-section measurements exist for these parent generator radioisotopes. The treatment of data and the evaluation methodology have to be the same as for single photon emitting radioisotopes (see below).

3. γ-Emitters

Discussion Leader: *Alex Hermanne*

In this discussion group the reactions leading to nuclides used in nearly every Nuclear Medicine department in the world were considered. With a few exceptions (^{77}Br and ^{211}At) several common characteristics can be found:

- the production makes use of medium energy proton on highly enriched targets;

- contaminating reactions limit the energy range in which practical production is possible;

- the end product is of great economical importance (mass production by commercial suppliers) and has led the IAEA to support starting up of production capabilities in several developing countries.

This last point makes the evaluation of available data and the generation of recommended values, that would be used worldwide, one of the main motivations of the CRP.

As data on excitation curves are probably also available in the "classified information" of the major radiopharmaceutical companies, the idea of trying to get hold of these data was discussed but rejected. Syed Qaim suggested that preference should be given to data that are widely available in the public domain and that private communications or non published data should be handled with care.

In view of the heavy burden laid on the theoretical evaluators it was decided to focus on the reactions leading to the main nuclide and not to include in general a study of the contaminating reactions. (Remark: this statement was altered later when it became clear that data available on more than one reaction channel for a given nuclide-bombarding particle combination would make evaluation and theoretical modelling more reliable).

It was also concluded that thick target yield calculations are only of second priority and should mainly be used as comparison base for selected benchmarking experiments where integral yields on thick targets are measured.

In discussing the commonly used radionuclides, the following remarks can be made:

(i) The production of ^{67}Ga depends on the available energy. Both (p,n) and (p,2n) routes should be evaluated, status of the experimental data for ^{67}Zn(p,n)^{67}Ga is good, but discrepancies in the value of the maximum exist for the 5 publications on the ^{68}Zn(p,2n)^{67}Ga reaction. Literature data have been compiled and analysed by the Debrecen group and recommended values will be published soon.

(ii) For ^{111}In production also three routes are used depending on the available energy. The three reactions, ^{111}Cd(p,n)^{111}In, ^{112}Cd(p,2n)^{111}In and ^{nat}Cd(p,x)^{111}In are well documented. No major problems exist in the data and compilation is underway in Debrecen.

(iii) The production of ^{201}Tℓ always proceeds along the classical ^{203}Tℓ(p,3n)^{201}Pb → ^{201}Tℓ route. A lot of data are available in the literature. Calculations have been performed by Shubin et al. The Debrecen group already started setting-up a data base and information is also grouped in Russia.

(iv) For the production of ^{81}Rb(^{81}Kr) generator only a few sets of measurements are available but no major discrepancies seem to exist. Evaluation should be possible.

(v) The ^{209}Bi(α,2n)^{111}At reaction data are reasonably well documented. Evaluation is possible.

(vi) The different reactions leading to ^{123}I have divergent status. The ^{124}Te(p,2n)^{123}I and ^{127}I(p,5n)^{123}Xe→^{123}I reactions are reasonably documented and a data set for evaluation could be prepared. A long discussion about the available data and their comparison with some ALICE calculations sprung up concerning the ^{123}Te(p,n)^{123}I reaction. At the moment only one set of measurements seem to be agreed on and no immediate action is possible. In the ^{124}Xe(p,x)^{123}I process several reactions contribute. The cross sections for the primary reactions have been measured after radiochemical separations in Jülich and non-destructively by Kurenkov et al. Although agreement between measurements is good for the ^{124}Xe(p,2n)^{123}Cs reaction, which contributes most to the final ^{123}I activity, a factor of nearly 2 difference exists between both the groups on the ^{124}Xe(p,pn)^{123}Xe reaction. More important is the discrepancy between directly measured ^{123}I yields where results of Kurenkov et al. and of the Brookhaven laboratory are much higher than those derived from the primary cross sections, while Jülich measurements confirm their cross section calculations.

Recommendations (in same order as discussion of reactions):

(i) The data sets for ^{67}Ga producing reactions are available for evaluation. Some additional experimental values have to be obtained on enriched targets for the ^{68}Zn(p,2n)^{67}Ga reaction near the maximum of the excitation function for normalisation purposes.

(ii) The data set for ^{111}In producing reactions can be prepared in the near future for evaluation.

(iii) The Brussels group should continue gathering published data for the $^{203}Tl(p,3n)^{201}Pb$ reaction and prepare an analysed and recommended data set. Comparison with calculated values and evaluation could be done by Yuri Shubin.

(iv) The existing data on the $^{82}Kr(p,2n)^{81}Rb$ reaction should be gathered, critically analysed and a recommended data set made available for evaluation.

(v) The same recommendations as above hold for the $^{209}Bi(\alpha,2n)^{211}At$ reaction.

(vi) For the $^{124}Te(p,2n)^{123}I$ and $^{127}I(p,5n)^{123}Xe$ reactions the available data should be gathered, critically analysed and prepared as a recommended data set. For the $^{123}Te(p,n)^{123}I$ reaction either new experimental data sets should be generated or a real evaluation has to be abandoned.

For the $^{124}Xe(p,x)^{123}I$ process four possible further actions are possible:

- average the existing experimental data on the two known primary excitation functions
- reassess the total ^{123}I yield under controlled irradiation conditions (at least 1 hour) and establish new data sets
- remeasure cross sections and yields
- look for possible "hidden" additional isomeric reactions and try to resolve the existing conflict between data

4. Medium Energy Data

Discussion Leader: *Stephen Mills*

Radioisotope production at medium energies offers the following advantages:

(i) A wider choice of production routes (and therefore, a greater variety of possible products, the possibility to find alternatives for difficult production routes - involving, for instance, enriched target material or complex chemistry - and, sometimes, a better end product);

(ii) higher production capacities, not only due to the inherently higher production yields (as a result of the lower stopping power), but also due to cumulative production (simultaneous production of more than one stable isotope in target material, production of short-lived precursor(s) in addition to the direct production of the radioisotope of interest, possibility to use tandem targets); and

(iii) more efficient target cooling (cooling at both front and rear surfaces of target(s) possible, since energy degradation in cooling water layers and, possibly, beam window foils can be accommodated, more favourable heated-volume-to-power ratio).

On the other hand, it also has some disadvantages:

(i) The use of enriched target material, if unavoidable, can become very expensive (more material required);

(ii) radiocontaminants are much more problematic;

(iii) enhanced radiation damage and activation; and

(iv) more difficult target handling and processing and increased radioactive waste.

After considering existing radioisotope production facilities with proton energies above 45 MeV and discussing the envisaged future availability of medium-energy, high-intensity commercial cyclotrons, the CRP decided that a proton energy of 100 MeV could be the upper limit for the scope of the present CRP, but that all existing relevant data above this energy should, nevertheless, be included in the compilations and evaluations eventually decided upon. This decision is also in line with the proposal for a 100 MeV machine for a "National Biomedical Tracer Facility" for the USA, currently under consideration.

The following production routes were identified as of particular interest in this energy region:

$Ge(p,x)^{67}Ga$
$Kr(p,x)^{81}Rb$
$Rb(p,x)^{82}Sr$
$In(p,x)^{111}Sn \rightarrow {}^{111}In$
${}^{127}I(p,5n)^{123}Xe \rightarrow {}^{123}I$
$Cu(p,x)^{62}Zn$
$Ga(p,x)^{68}Ge$

All the radioisotopes produced in these reactions have already been singled out in the earlier sessions on "Positron Emitters" and "γ-Emitters".

It was also emphasized that, in contrast to the lower energy production routes, it is essential also to include the production routes of all possible radiocontaminants in these cases in the eventual data base. It was furthermore recommended that all proton beam monitor excitation functions included in the CRP should also be considered up to medium energies.

Finally, it was the view of the meeting, in the light of the very limited number of accelerators with sufficient beam intensities involved in the production of radioisotopes by means of d, ^{3}He or α beams, that the scope of the present CRP should not extend to medium energies (above 30-40 MeV) in the case of these particles.

5. Calculation and Analysis of Excitation Functions

Discussion Leader: *Yuri Shubin*

A number of presentations outlined the range of accelerator production of radioisotopes for medical applications. Yet there appears the wish to understand experimental reaction cross sections in terms of nuclear model calculations. It is possible to use these models as a guide in selecting the best experimental conditions for producing a given radioisotope without having to measure all the reactions of interest.

Zhuang Youxiang (CNDC) reported on some efforts to develop computer codes based on statistical and preequilibrium models. These codes have been used to interpolate and extrapolate experimental data for medium mass and fissile nuclei.

S. Mills (NAC, South Africa) presented results of calculations for the description of the data measured at NAC, Faure. The ALICE-85 code has been used for these calculations. The results are in a reasonable agreement with the data for multiparticle emission reactions in many cases.

Y. Shubin presented the results of calculations and analyses for a number of reactions, mostly proton and light charged particle induced reactions at energies up to 100 MeV using various codes (STAPRE, ALICE, GROGI). It was noted that there exist several versions of ALICE code that do not take into account some of the important physical effects, such as gamma-emission competition, preequilibrium emission of clusters, nuclear structure effects in the nuclear level density etc. It was recommended to use more sophisticated codes, like STAPRE, especially near thresholds.

6. Evaluation Methodology

Discussion Leader: *Pavel Obložinský*

It was pointed out that the evaluation methodology for charged particle reaction cross sections is not yet well established. A useful paper, by the Chinese group, was contributed to the 1992 Workshop on Nuclear Data Methodology in Brookhaven. Based on a long-term experience from evaluations of neutron reaction cross sections it is accepted that an evaluator should have an expertise in nuclear experimentation and instrumentation, nuclear reaction theory, modelling and calculations, mathematical statistics, programming techniques and applications of nuclear data.

Two basic evaluation procedures were discussed. An experimental procedure is based purely on measurements, compilation and analysis of experimental data. This method is valuable especially in cases where theoretical approaches are not well established or they are difficult to apply. This applies particularly to reactions on light nuclei such as those used to produce positron emitters for PET applications.

A theoretical procedure involves a strong component of nuclear reaction calculations, comparison and fitting to experimental data. This method should be preferred whenever possible. It is especially suitable in medium and heavy nuclei where physics of nuclear reactions is well established. Comprehensive experimental information should be taken into account, if available. However, it was pointed out that requirements on nuclear data for medical RI production differ substantially from those for neutron energy applications. Therefore, generally of most concern is the actual production reaction.

Suitable codes were discussed. ALICE was found to be very useful, although some participants doubted its applicability at incident energies near reaction thresholds. In this case much more sophisticated codes should be preferred. It is recognized that much more experience with charged particle reaction evaluations is needed. The evaluation procedures are subject to further refinements and improvements.

Evaluation methodology based purely on experimental data was summarized by **Zhuang Youxiang** into the following 3 steps:

The first step is *Compilation and Collection of Data.* The measured data need to be compiled in EXFOR format for preserving, exchanging and using the related experimental data. The compilation should be as complete as possible.

The second step is *Analysis of Data.* Compiled experimental data are analysed on the basis of measuring method and instrumentation, correction factors and covariance matrices etc. Based on this, final selection of experimental data is done. All chosen data are plotted in different figures. If the discrepancy among them is large, a further and refined analysis is needed to decide which should be accepted or rejected.

The third, final step is *Evaluation of Data.* The data fit programs are used to obtain the evaluated cross sections or equal-weight average can be adopted. In favourable cases also the covariances can be obtained.

Concerning nuclear reaction theory and model calculation, Zhuang Youxiang noted that the present power of appropriately parameterised nuclear reaction theories and nuclear model calculations includes inter- and extra-polation of experimental results, prediction of unknown nuclear data, checking the internal consistency between measurements and a guide in deciding among discrepant experimental results.

The Chinese experience shows that if there is a need for the data of a specific reaction channel but not for a complete set of all the partial reaction channels, one should do the following:

(1) If there are accurate and enough experimental data, the recommended values are taken from evaluated ones

(2) If experimental data are scarce, theoretical model calculation or systematics can represent them well, model or systematics calculation can be accepted.

7. Recommendations

(1) The calculation of excitation functions is a valuable tool in experimental programmes for the production of radioisotopes for medical applications. The IAEA should therefore provide all the necessary assistance in selecting and making such codes available.

(2) Recent developments in nuclear reaction theories and nuclear structure information should be incorporated in existing codes. In this regard the Obninsk version of ALICE, namely ALICE-IPPE can be recommended. Y. Shubin is therefore requested to make his code available in user-friendly form to individual experimentalists as well as the IAEA as soon as possible. Other theoretical codes should, however, also be employed within the CRP as far as possible.

(3) A major benchmarking exercise of codes for activation charged particle reaction cross section calculations is underway at NEA (P. Nagel and R. Michel). The CRP should be cognizant of results of this exercise and consider its conclusions at the next CRP meeting.

(4) Guidelines for the proper and accurate measurement of excitation functions should be drawn up and published. S.M. Qaim and F. Tárkányi will give attention to this as soon as practically possible.

(5) Guidelines for the proper use of the evaluated data should be drawn up and made available together with these data. S.M. Qaim and F. Tárkányi will also give attention to this.

(6) As wide a variety of theoretical codes as possible should be employed in the evaluation of the experimental data sets.

(7) The CRP should ensure that all data used in the programme are taken up in the EXFOR file. Action: F. Tárkányi.

(8) The second Research Co-ordination Meeting should be held preferably in March/ April 1997.

V. Work Plan

The following work plan for the next 18 months (i.e. until the next CRP Meeting) has been agreed upon:

1. Evaluation of Monitor Excitation Functions

Since monitor excitation functions form the basis of all experimental cross-section measurements, the highest priority should be given to the evaluation of monitor reactions of primary interest, namely those needed for proton beam monitoring.

The following division of responsibilities was agreed upon:

ATOMKI Debrecen:	$^{nat}Ti(p,x)^{48}V$
VU Brussels:	$^{nat}Ni(p,x)^{57}Ni$ *)
KFA Jülich:	$^{nat}Cu(p,x)^{62}Zn$ $^{nat}Cu(p,x)^{63}Zn$ $^{nat}Cu(p,x)^{65}Zn$
NAC Faure:	$^{27}A\ell(p,3p3n)^{22}Na$ $^{nat}Cu(p,x)^{56}Co$

*) Note: It was pointed out by S. Mills later after the meeting that ^{57}Ni activation may be affected by secondary neutrons due to $^{58}Ni(n,2n)$ reactions. VU Brussels should consider this effect in their analysis, removing eventually ^{57}Ni from the present list.

Procedures:

(i) F. Tárkányi to act as a coordinator for the experimental groups and to supply final collected experimental data to China Nuclear Data Centre, IPPE Obninsk and Lawrence Livermore National Laboratory for evaluation.

(ii) All experimental groups to contribute all available information at their disposal on each of the above reactions to the relevant responsible group as soon as possible.

(iii) All experimental groups to screen available experimental data for possible errors, shortcomings, etc.

(iv) Copies of all correspondence on the above to be supplied to the Scientific Secretary of the CRP (P. Obložinský).

(v) Format for information exchange: Copies of all relevant publications; tables to be supplied in DOS Text Format and in Excel 5.0.

Deadlines:

Collected experimental data to be supplied by F. Tárkányi to evaluating groups before 1 June 1996. Evaluating groups to attempt to complete evaluations before next CRP meeting in 18 months.

2. Evaluation of Excitation Functions of γ-Emitters

The evaluation of the highest priority γ-emitters should proceed in parallel with those of the monitor excitation functions in 1. above. The following division of responsibilities in this regard was agreed upon:

ATOMKI Debrecen:	$^{111}Cd(p,n)^{111}In$ $^{112}Cd(p,2n)^{111}In$ $^{nat}Cd(p,x)^{111}In$
VU Brussels:	$^{203}T\ell(p,3n)^{201}Pb \rightarrow {}^{201}T\ell$
KFA Jülich:	$^{124}Te(p,2n)^{123}I$
NAC Faure:	$^{127}I(p,5n)^{123}Xe \rightarrow {}^{123}I$

Procedures: As in 1. above

Deadlines: As in 1. above

3. Experimental Measurements

The following experimental measurements should be completed within the next 18 months (i.e. before the next CRP Meeting):

ATOMKI Debrecen and VU Brussels and KFA Jülich	$^{nat}Ti(d,xn)^{48}V$ from threshold up to 22 MeV (Insufficient exp. data)
ATOMKI Debrecen and VU Brussels	$^{68}Zn(p,2n)^{67}Ga$, possibly on enriched target (Discrepancies in available exp. data) Target date: Sept. 1997
ATOMKI Debrecen and KFA Jülich	$^{18}O(p,n)^{18}F$ (Measurement of excitation function with moderate beam energy resolution)

Appendix 1

First Research Co-ordination Meeting on
"Development of Reference Charged Particle Cross Section Data Base for Medical Radioisotope Production"

IAEA Headquarters, Vienna, Austria
15 to 17 November 1995

Meeting Rooms B07-42 and A24-13

Scientific Secretary: P. Obložinský

AGENDA

Wednesday, 15 November 1995

08:00 - 08:45 Registration of Participants
(IAEA Information Desk, Tower C, Ground Floor)

09:00 - 12:30 **Joint Session with the Consultants' Meeting on "Automated Synthetic Systems"** (Meeting Room B07-42)

Opening (P. Obložinský and H. Vera Ruiz - IAEA)
Election of Chairman
Adoption of Agenda

S.M. Qaim (Germany)
- Lecture:
Status of nuclear data for cyclotron production of medical radioisotopes, with particular reference to longer-lived β^+ emitters, therapeutic radionuclides and generator systems

R.M. Lambrecht (USA)
- Lecture:
Status of nuclear data for cyclotron production of single photon emitters, with particular reference to ^{99m}Tc and ^{123}I

P. Cohilis (Belgium)
- Lecture:
High intensity cyclotrons for radioisotope production

All Participants
- Discussion:
Status of medically oriented cyclotrons, new projects

14:00 - 15:30 **Laboratory Reports** (Meeting Room A24-13)

- Belgium, China, Germany, Hungary, Russia, South Africa (each 10 min)

16:00 - 17:30 **Monitor Reactions**

Discussion Leader: F. Tárkányi

- Short contributions, general discussion, recommendations for the CRP

17:30 **Welcome Drink** (in front of NDS Library, A23-40)

Thursday, 16 November 1995

09:00 - 10:30 **Positron Emitters**

Discussion Leader: S.M. Qaim

- Short contributions, general discussion, recommendations for the CRP

11:00 - 12:30 **γ-Emitters**

Discussion Leader: A. Hermanne

- Short contributions, general discussion, recommendations for the CRP

14:00 - 15:30 **Intermediate Energy Data**

Discussion Leader: S.J. Mills

- Short contributions, general discussion, recommendations for the CRP

16:00 - 17:30 **Theory and Model Calculations**

Discussion Leaders: Y.N. Shubin and P. Obložinský

Y.N. Shubin
- Lecture:
  Nuclear model calculations applied to medical radioisotope production

- Short contributions, general discussion, recommendations for the CRP

19:00 **Dinner**, "Gösser Bierklinik", Steindlgasse 4, Wien (near Hotel Wandl)

Friday, 17 November 1995

09:00 - 10:30 **Evaluation Methodology**

Discussion Leaders: Zhuang Youxiang and P. Obložinský

- Short contributions, general discussion, recommendations for the CRP

11:00 - 12:30 **Compilation of the Recommendations and Work Plan of the CRP**

- Briefing on results of the Consultants' Meeting (S.M. Qaim and H. Vera Ruiz)
- Recommendations
- Work Plan

14:00 - 16:00 **Final Discussion, Draft Report**

First Research Co-ordination Meeting on
"Development of Reference Charged Particle Cross Section Data Base for Medical Radioisotope Production"

IAEA Headquarters, Vienna, Austria
15 to 17 November 1995

Scientific Secretary: P. Obložinský

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Information Sheet on the Research Coordinated Programme
" Development of Reference Charged Particle Cross Section Data Base for Medical Radioisotope Production"

1. Scientific Background

Charged particle induced nuclear reaction cross sections are of interest both from the viewpoint of theory and applications. Of considerable significance are the excitation functions obtained via the activation technique. Those data find applications in several fields

- radioisotope production, primarily for medical applications (but also for industrial and agricultural use)
- monitoring of light charged particle beams (p, d, ^3He, α) in cyclotron facilities
- surface analysis in industrial applications
- astrophysics and cosmochemistry.

Primarily, data in the low energy range up to 30 MeV are required. Much experimental data already exist. But for production of certain radioisotopes higher energy data, up to about 60–80 MeV, are gaining importance.

The use of nuclear theory for reliable prediction of cross section data is still limited. Experimental studies and development of models for improvements in calculational capabilities are needed.

During the past twenty years, many laboratories have reported a large body of experimental data, and the charged-particle data centers have compiled most of those data in EXFOR form. However, no international effort has been devoted hitherto to the standardization of those data. Some of the national centers interested in this type of work (NNDC, USA; KaChaPag, Germany; CAJAD, Russia; RIKEN, Japan) never started evaluation work, and the NEA Data Bank does not have the mandate to initiate charged particle activity. The IAEA has thus the unique opportunity and possibility to undertake this responsibility.

2. Supporting Information

The subject was discussed critically in several review articles

- S.M. Qaim, Radiochimica Acta **30**, 147-162 (1982); ibid **41**, 111-117 (1987)
- Hashizume, in Proc. Int. Conf. Nuclear Data for Science and Technology, Mito, 1988 (Saikon, Tokyo, 1988) pp. 1067-1072
- Stöcklin, in Proc. Int. Conf. Nuclear Data for Science and Technology, Jülich, 1991 (Ed. S.M. Qaim, Springer Verlag, Berlin, 1992) pp. 573-578

and at three IAEA meetings

- Report INDC(NDS)-123 "Consultants' Meeting on Nuclear Data for Medical Radioisotope Production" (IAEA Vienna, April 1981)

- Report INDC(NDS)-195 "Consultants' Meeting on Data Requirements for Medical Radioisotope Production", Tokyo, 24–28 April 1987 (Ed. K. Okamoto, IAEA Vienna, 1988)
- Report INDC(NDS)-245 "Advisory Group Meeting on Intermediate Energy Data for Applications" (IAEA Vienna, October 1990)

Based on the recommendations made in those meetings a modest attempt was made to collect the available information on the Monitor Reactions

- O. Schwerer and K. Okamoto: "Status Report on Cross–Sections of Monitor Reactions for Radioisotope Production", Report INDC(NDS)-218 (IAEA Vienna, 1989)

No action was taken so far on the other recommendations.

A large number of medically oriented cyclotrons have been running in North America, Western Europe and Japan for more than two decades. In recent years, 30-40 MeV cyclotrons have been installed in several other countries (e.g. Australia, Argentina, Taiwan, PR China (Shanghai and Beijing), South Korea, Iran, Indonesia, etc.) and smaller cyclotrons (E_p < 20 MeV) have been purchased or ordered by North Korea and Egypt. All those countries will soon start producing standard γ–emitting radioisotopes like ^{67}Ga,^{111}In,^{201}Tl and ^{123}I, commonly employed in diagnostic investigations using γ–cameras and Single Photon Emission Tomographs (SPET). Although the production methods are well established, there are no evaluated and recommended data sets available. The need for standardization is thus imminent.

3. Scientific Scope and Programme Goals

The CRP should cover the following areas:

- Compilation of data on the most important reactions for monitoring light ion charged particle beams (p, d, ^3He, α). Evaluation of the available data (both by fitting and theory). Produce recommended curves.
- Compilation of production cross section data on radioisotopes most commonly used in medicine. Evaluation of the data. Produce recommended curves.
- Validation of the evaluated curves through integral measurements (thick target yield measurements at low beam currents).
- New measurements in cases where data base is weak or where evaluations reveal deficiencies.
- Development of calculational tools for predicting unknown data.

Although the major emphasis in the CRP will lie in the energy region up to 30 MeV, higher energy data up to about 60–80 MeV will also be considered. It is realized that the evaluation methodology for charged particle data is not well developed and some teach-in effort may initially be necessary. The final objective of the CRP should be to produce a recommended data base and a Handbook, covering reactions used for monitoring beam currents and for routine production of medically important radioisotopes. Part of the results will be used in the International Reference Activation Data Library being developed under another CRP.

Appendix 3

Consultants' Meeting on

Automated Synthesis Systems for the Cyclotron Production of ^{18}F and ^{123}I and their labelled Radiopharmaceuticals

IAEA Headquarters, Vienna
13 - 16 November 1995

Scientific Secretary: H. Vera-Ruiz

Summary and Recommendations

1. The consultants reaffirmed the importance of Fluorine-18 and Iodine-123 in radiopharmaceutical development for molecular nuclear medicine. These tracers should play an increasing role in all countries with cyclotrons.

2. For radionuclide production, use of [^{18}O]water targets and enriched tellurium targets for the production of Fluorine-18 and Iodine-123, respectively, is recommended for smaller cyclotron facilities. For large scale production of ultra high purity ^{123}I, the use of ^{124}Xe enriched target should be considered.

3. The implementation of methodologies for no-carrier-added radioiodination and radiofluorination is recommended. Automated or semi-automated systems should be considered where feasible.

4. Among the Fluorine-18 labelled radiopharmaceuticals, 2-[^{18}F]FDG plays a major role. In the case of Iodine-123, there are several important radiotracers: β-CIT, IBZM, iomazenil, IDEX, IMT, and BMIPP.

5. As an important research area, the development of Fluorine-18 and Iodine-123 labelled peptides for applications in oncology, cardiology, neurology and infectious disease is recommended.

6. It is recognized that tracer validation, including *in vitro* cell binding and *in vivo* studies, is a vital aspect of radiopharmaceutical development.

7. It is recommended that a CRP entitled "The Development and Quality Control of Fluorine-18 and Iodine-123 Labelled Peptides and their Evaluation" be supported. A Research Project outline has been worked out.

8. The concept of imaging endo radiotherapy agents via gamma or positron-emitting isotopes or analogues using SPET or PET is recognized.

9. It is recommended that the 1983 Cyclotron Directory should be updated in 1996. The recommended format questionnaire has been prepared.

10. A Consultants Meeting on emerging specific alternative technologies versus existing methods for ^{99}Mo production and ^{99m}Tc production is recommended.

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