

Summary

Radionuclides have been playing an important role both in energy and non-energy related applications. In recent years their significance in medical diagnosis and therapy has considerably increased. In particular, appreciable efforts are being devoted to their use in internal radionuclide therapy. For this purpose radionuclides emitting low-energy but highly ionizing radiation (i.e. α -particle, low-energy β^- particles, Auger electrons, X-rays, etc.) are in great demand. Those radionuclides are often produced at reactors and cyclotrons. In reactor irradiations neutron induced reactions occur whereas at cyclotrons charged particle induced reactions are dominant. This study mainly concentrates on charged particle induced reactions for the production of three therapeutic radionuclides, namely, ^{103}Pd , ^{186}Re and ^{67}Cu . The aim was to establish standardized reliable data which could be used to deliver high-quality products suitable for medical applications.

For the compilation of data, the major source was the EXFOR file of the IAEA. In addition, some original literature data were collected from international journals as well as from four most active laboratories, namely, the *Institut für Nuklearchemie, Forschungszentrum Jülich GmbH, Jülich, Germany*, the *Institute of Experimental Physics, University of Debrecen, Debrecen, Hungary*, the *Institute of Nuclear Research of the Hungarian Academy of Sciences (ATOMKI), Debrecen, Hungary* and *Cyclotron Department, Vrije Universiteit Brussel, Laarbeeklaan, Brussels, Belgium*. Together with their cooperation partners in Russia, Japan, South Africa and USA they cover the full spectrum of experimental studies in this field. It is believed that no major work has been left out in this study.

For the normalization of data, the monitor reaction cross sections recommended in the IAEA-TECDOC-1211 [15] were taken as standard and for decay data NUDAT 2.4 [70] library of BNL, USA was considered as a reference. In the case of ^{186}Re , the decay data used in each experiment were compared with the precise gamma ray emission probabilities recently reported by Miyahara et al.(2000) [109].

For consistency checks in the experimental data, nuclear model calculations were done by using compound-precompound codes ALICE-IPPE, STAPRE, EMPIRE and TALYS. The ALICE-IPPE calculations were done at the *Government College University, Lahore* while the rest of the calculations were done at the *Institute of Experimental Physics, University of Debrecen, Debrecen, Hungary*. Some of the EMPIRE calculations were done at the *IAEA-headquarters, Vienna, Austria*. For each production reaction of an individual radionuclide, the ratio of the measured/model calculated cross section was developed. The ratio was then fitted using a polynomial function with weighting errors to estimate the energy dependent normalization factor. It was assumed that cross sections are functions of model calculations and the energy dependent factor.

To estimate the best fit, we neglected the data that were out of 3σ limit of the uncertainties of the polynomial fit. After neglecting those data, polynomial fittings were done again by taking into account the weighting factor of the uncertainties. The normalization factor for each model calculation was estimated and multiplied by the model calculation. 95% confidence limits were selected to estimate the uncertainty in the recommended data. This technique allowed us to remove major energy dependence of the cross section (not completely because the polynomial fits of the ratios were not simple linear functions of the first order). Recommended sets of the data were generated by taking average and average fit of the three normalized model calculations. Those values are tabulated for ready use. For a few reactions under study, the evaluated curves were compared with the available data of an ongoing CRP of the IAEA. Many other reactions, however, have been evaluated for the first time.

The recommended cross sections were finally utilized to calculate integral yields of the products. Those data are believed to be more accurate than the values calculated from measurements of individual authors. The cross section data and calculated yields were finally used to determine the optimum conditions for the production of the three investigated radionuclides.

A calculation of all the reaction yields for the ^{103}Pd shows that the $^{\text{nat}}\text{Ag}(p,x)^{103}\text{Pd}$ process at about 70 MeV gives the highest yield but the level of ^{100}Pd impurity amounts to about 50% [cf. 75] and a waiting time of about 25 days may be necessary to decrease its contribution to $< 1\%$ of

^{103}Pd . Therefore, higher incident proton energies should be avoided to keep the level of ^{100}Pd as low as possible. Thus, low energy cyclotron production via the $^{103}\text{Rh}(p,n)^{103}\text{Pd}$ or the $^{103}\text{Rh}(d,2n)^{103}\text{Pd}$ reaction is more suitable. At present, low and medium energy commercial cyclotrons deliver proton beams of much higher intensity than deuteron beams, so the $^{103}\text{Rh}(p,n)^{103}\text{Pd}$ reaction is the method of choice.

The study of the yields of ^{186}Re reflected that, for the same energy range, the $^{186}\text{W}(d,2n)^{186}\text{Re}$ reaction leads to much higher yield than the $^{186}\text{W}(p,n)^{186}\text{Re}$ reaction [129]. But, the available commercial production cyclotrons have deuteron energy only half of the proton energy (e.g. 18 MeV p/9 MeV d; 30 MeV p/15 MeV d) so that the achievable ^{186}Re yield via the (d,2n) reaction is considerably reduced. Also, the intensity of proton beams is much higher than the presently available deuteron beams. In consideration of radionuclidic purity of ^{186}Re , the use of highly enriched ^{186}W target for production is recommended. For the optimization of production route, 18 MeV proton energy is suggested, although the yield of ^{186}Re is further increasing. It is due to the formation of ^{184g}Re ($T_{1/2} = 38.0$ d) via the $^{186}\text{W}(p,3n)$ – reaction. Above 18 MeV, even if highly enriched ^{186}W is used as target material, the amount of ^{184g}Re formed would be significant [118]. This increases the impurity level in ^{186}Re . Therefore, the method of choice is the $^{186}\text{W}(p,n)^{186}\text{Re}$ reaction at about 18 MeV.

The production of the medical radionuclide ^{67}Cu is possible via the reactions $^{64}\text{Ni}(\alpha,p)^{67}\text{Cu}$, $^{68}\text{Zn}(p,2p)^{67}\text{Cu}$ and $^{70}\text{Zn}(p,\alpha)^{67}\text{Cu}$. The corresponding particle energies and evaluated reaction cross section values are mentioned. The production possibility in different energy regions via different nuclear reactions has been clarified. The study of all reactions for the production of ^{67}Cu leads to the conclusion that the $^{68}\text{Zn}(p,2p)^{67}\text{Cu}$ reaction should be the method of choice if high energy proton beams would be available. But the proton irradiation of enriched ^{68}Zn gives large amounts of ^{67}Ga along with ^{67}Cu . Both the ^{67}Cu and ^{67}Ga have almost identical gamma-ray spectrum so a very clean separation of ^{67}Cu is necessary. Another important radioactive impurity is the ^{64}Cu , which is formed in the investigated energy regions. However, ^{64}Cu has a shorter half-life (12.7 h), and decays earlier than the ^{67}Cu . The $^{70}\text{Zn}(p,\alpha)^{67}\text{Cu}$ reaction is more favourable if cyclotrons of low energy are available, in this case the ^{67}Cu yield will be rather low. The other drawback is the cost of enriched ^{70}Zn target that is rather high. The major advantage of this route

is the radiochemical purity of the product. The least important is the $^{64}\text{Ni}(\alpha,p)^{67}\text{Cu}$ process. It gives low yield of ^{67}Cu and the cost of the enriched target material is very high.