Residual Nuclide Production From Iron, Lead, And Uranium By Neutron-Induced Reactions Up To 180 MeV

R. Michel[1], W. Glasser[1], U. Herpers[2], H. Schuhmacher[3], H.J. Brede[3], V. Dangendorf[3], R. Nolte[3], P. Malmborg[4], A.V. Prokofiev[5,6], A.N. Smirnov[6], I. Rishkov[6], D. Kollár[7], J.P. Meulders[8], M. Duijvestijn[9], A. Koning[9]

1 Center for Radiation Protection and Radioecology, University Hanover, Germany
2 Dept. for Nuclear Chemistry, University of Cologne, Germany
3 Physikalisch-Technische Bundesanstalt (PTB), Braunschweig, Germany
4 The Svedberg Laboratory, University of Uppsala, Sweden
5 Department of Neutron Research, University of Uppsala, Sweden,
6 The Khlopin Radium Institute, St. Petersburg, Russia
7 Dept. of Nuclear Physics, Comenius University, Bratislava, Slovak Republic
8 Université Catholique de Louvain (UCL), Louvain-la-Neuve, Belgium
9 Nuclear Research and Consultancy Group, Petten, The Netherlands

Abstract. Within the HINDAS project, activation experiments with quasi mono-energetic were performed at UCL and TSL. Cross sections for the production of residual radionuclides were derived from the measured activities by unfolding based on the neutron spectra inside the target stacks and starting from “guess” excitation functions. Exemplary results are presented and compared with theoretical calculations using the TALYS code.

INTRODUCTION

Within the HINDAS (*High and Intermediate Energy Nuclear Data for Accelerator-Driven Systems*) project [1], total of 10 activation experiments with quasi mono-energetic neutrons produced by the ^{7}Li(p,n)^{7}Be reaction with proton-energies between 36.4 and 178.8 MeV were performed at the neutron beam lines at The Svedberg Laboratory (TSL), University Uppsala, Sweden and the Universite Catholique de Louvain La Neuve (UCL), Belgium in order to determine excitation functions for the production of residual radionuclides from a variety of target elements. In a previous report [2], the experimental techniques and the methods of evaluation were described and detailed references were given. Here, we report on the progress of this work, present results for the target elements Cu, Fe, Pb, and U, and compare them with theoretical predictions using the TALYS code which was developed within the HINDAS project (Koning et al., these proceedings).

METHODOLOGY

Cross sections cannot be directly calculated from the production rates P (equ. 1) which are determined from γ-spectrometric measurements since the neutrons used are just "quasi mono-energetic" with only about 30 to 50% of the neutrons in the high-energy peak with a width of a few MeV.

$$P = \frac{N_L}{A_T}\int s\,(E)\cdot J(E)\,\mathrm{d}E \text{ with } J(E) = \frac{\mathrm{d}^2\Phi(E)}{\mathrm{d}E\,\mathrm{d}t} \quad (1)$$

Therefore, the neutron cross sections have to be extracted from the experimentally determined production rates in a series of irradiation experiments with different neutron energies by unfolding. To this end, we used the STAY’SL formalism [3-4] starting from guess functions calculated by the ALICE-IPPE code [5]. Information on the energy dependence of the neutron spectra $J(E)$ in the targets was obtained by

modeling the neutron spectra by Monte Carlo techniques; see [2] for details and references. These transport calculations started either from the experimentally determined neutron spectra (at UCL) or from the systematic of experimentally measured neutron emission spectra of the ^{7}Li(p,n)-reaction. The calculations described the transport of the neutrons into the target stacks and into the individual targets as well the production and transport of secondary particles inside the massive target stacks which cannot be neglected.

In our previous work [2], an iterative approach of Kim et al. [6] was used which does not give entire excitation functions but resulted in individual cross sections at the peak neutron energies of the different experiments, only. We now succeeded in a complete unfolding of the excitation functions. It turned out that the results are rather independent of the guess functions used. The unfolding gives also complete standard uncertainties of the cross sections for all the energies. A further advantage is that this procedure can be repeated and the results can be improved if new information becomes available.

RESULTS AND DISCUSSION

We present here exemplarily results for the production of ^{56}Co from natural copper (Fig. 1), of ^{54}Mn, $^{52m+g}$Mn, ^{51}Cr, and ^{48}V from natural iron (Fig. 2), of ^{200}Pb, ^{196}Au, and ^{103}Ru from natural lead and of ^{103}Ru from natural uranium (Fig. 3). For each product nuclide, we present also cross sections for its proton-induced production to allow a comparison of the different modes of formation. For some of the excitation functions, we give also the guess functions used. Further, the new data are compared with theoretical predictions calculated by the TALYS code.

Only for the reaction ^{nat}Cu(n,X)^{56}Co (Fig. 1) exist results from other authors. The agreement between our excitation function and the cross sections for neutron-energies of 55 MeV, 61 MeV, and 75 MeV reported by Kim et al. [6] is quite satisfying. This reaction shows significant differences between the p- and n-induced productions of ^{56}Co over the entire energy range. Above

For the target element iron (Fig. 2), the differences between the p- and n-induced production modes are clearly seen for $^{52m+g}$Mn for all energies and for ^{54}Mn and ^{48}V at energies below 50 and 70 MeV, respectively. These differences can be attributed to individual reaction channels involving differently light complex particles. For ^{51}Cr and ^{46}Sc (not shown) these differences are marginal.

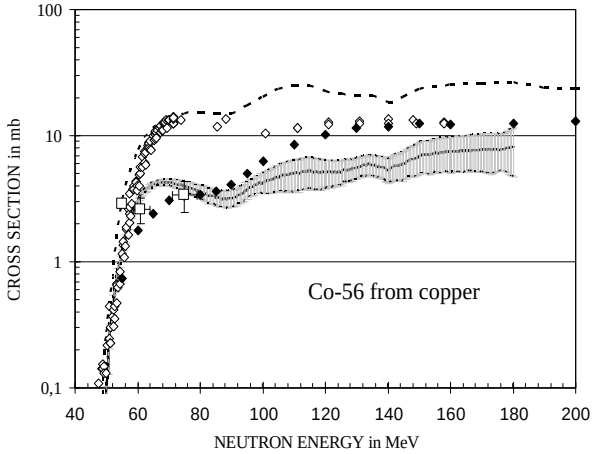

FIGURE 1. Excitation functions for the production of ^{56}Co from natural copper by n- and p-induced reactions. For n-induced reactions the STAY'SL results and their uncertainties are given as solid line and shaded area, the ALICE-IPPE guess function as dotted line and the TALYS calculations as full diamonds. Open diamonds are experimental cross sections for ^{nat}Cu(p,X)^{56}Co [7]. Open squares are experimental results for ^{nat}Cu(n,X)^{56}Co [6].

The rather large uncertainties in the excitation function for the production of ^{48}V from iron for energies around 130 MeV are due to problems with the low-level counting of the small activities produced. A similar problem is to be seen for the production of ^{200}Pb from lead (Fig. 3) at 110 MeV where a minimum in the excitation function occurs without physical reason.

For the target element lead (Fig. 3), the differences between p- and n-induced production modes are due to hindrance of p-evaporation in heavy target elements (^{196}Au and ^{200}Pb) and to differences in the general probability of p- and n-induced fission (^{103}Ru). The differences seen between p- and n-induced fission of lead do not show up for ^{99}Mo from uranium (Fig. 3).

The structures in the excitation function for ^{103}Ru from uranium predicted by the TALYS code at energies below 30 MeV cannot be resolved by the present experimental technique. Generally, the results of the TALYS calculations reproduce the experimental data fairly well though some problems pertain. It is interesting to not that there are significant differences between the TALYS and the ALICE-IPPE calculations in several cases.

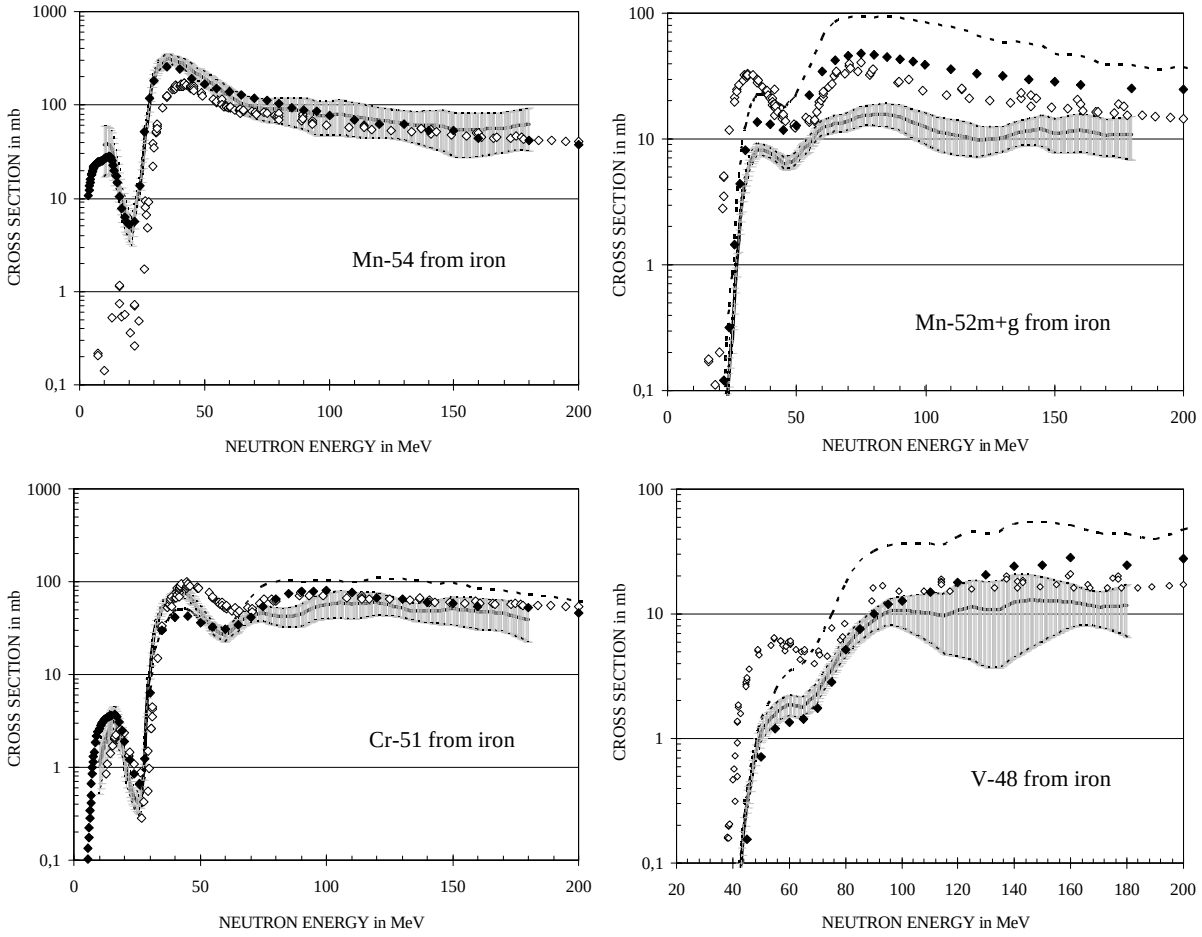

FIGURE 2. Excitation functions for the production of ^{54}Mn, $^{52m+g}$Mn, ^{51}Cr and ^{48}V from natural iron by n- and p-induced reactions. For n-induced reactions the STAY'SL results and their uncertainties are given as solid line and shaded area, the ALICE-IPPE guess function as dotted line and the TALYS calculations as full diamonds. Open diamonds are cross sections for the p-induced production from [7] and Michel et al. (these proceedings).

CONCLUSION

The results of these studies demonstrate that there are distinct differences between n- and p-induced production modes of residual nuclides which are of importance for accelerator-driven technologies and for other applications. The experimental determination of the respective excitation functions is feasible using irradiation experiments at high-current sources of quasi-monoenergetic neutrons and unfolding the underlying excitation functions from the experimental production rates.

Further excitation functions for the n-induced production of residual nuclides are needed to validate theoretical predictions of cross sections for their production and to improve the theoretical understanding of the differences between n- and p-induced reaction modes.

ACKNOWLEDGMENTS

The authors wish to thank the authorities of UCL and TSL for the beam-time and the accelerator staff for their kind co-operation. This work was funded within the EC 6th Framework project HINDAS.

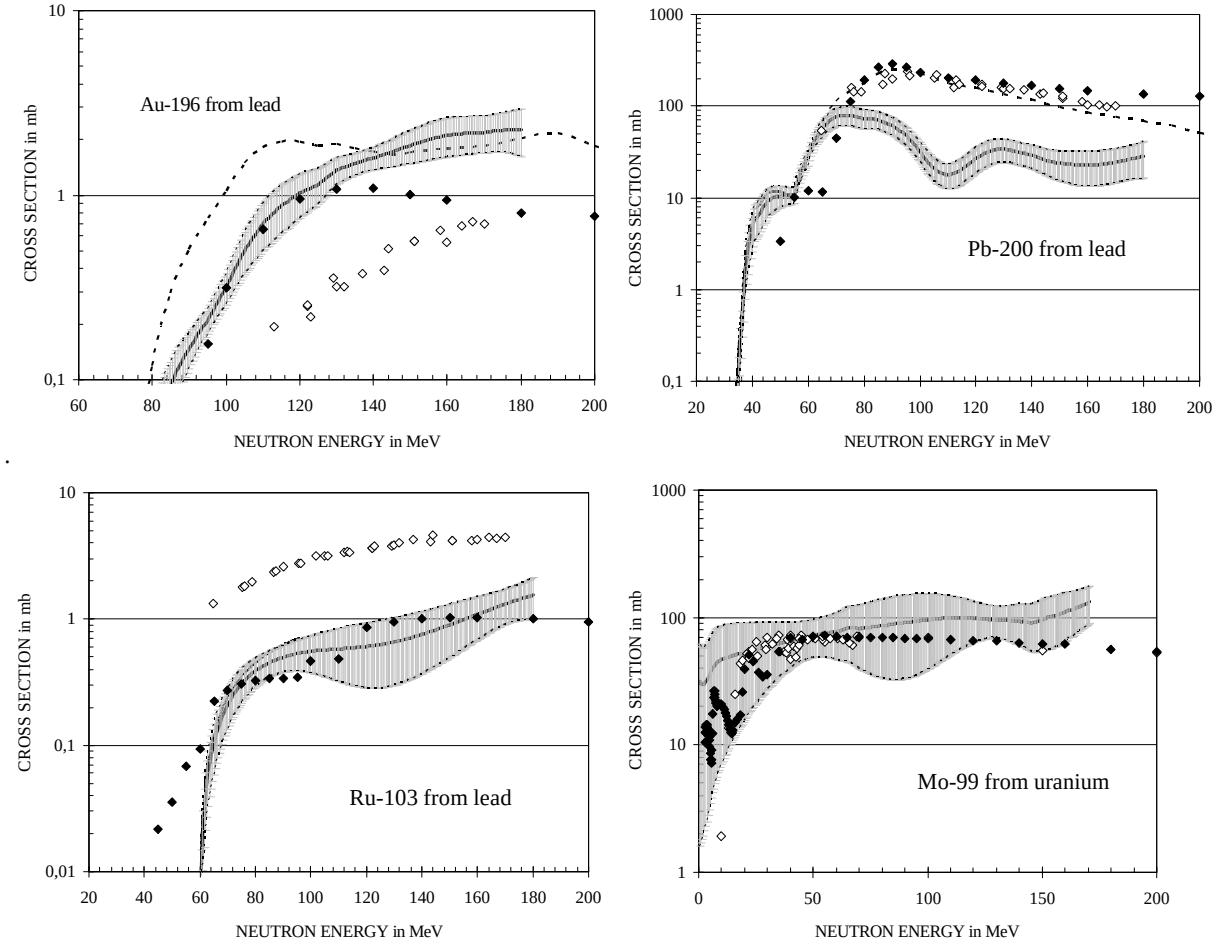

Figure 3. Excitation functions for the production of ^{196}Au, ^{200}Pb, and ^{103}Ru from natural lead and for ^{99}Mo from natural uranium by n- and p-induced reactions. For n-induced reactions the STAY'SL results and their uncertainties are given as solid line and shaded area, the ALICE-IPPE guess function as dotted line and the TALYS calculations as full diamonds. Open diamonds are cross sections for the p-induced production from [8].

REFERENCES

1. Koning, A., Beijers, H., Benlliure, J., Bersillion, O., Cugnon, J., Duijvenstein, M., Eudes, Ph., Filges, D., Haddad, F., Lebrun, C., Lecolley, F.-R., Leray, S., Meulders, J.-P., Michel, R., Neef, R., Nolte, R., Olsson, N., Ramström, E., Schmidt, K.-H., Schuhmacher, H., Slypen, I., Synal, H.-A., and Weinreich, R., Nucl. Science Technology, Supplement **2**, 1161-1166 (2002).
2. Glasser, W., Michel, R., Neumann, S., Schuhmacher, H., Brede, H.J., Dangendorf, V., Nolte, R., Herpers, U., Smirnov, A.N., Ryzhov, I., Prokofiev, A.V., Malmborg, P., Kollár, D., and Meulders, J.P., Nucl. Science Technology, Supplement **2,** 373 - 376 (2002).
3. Perey, F.G., *Least-squares dosimetry unfolding: The program STAY'SL*, Rep. ORNL-TM-6062, revised by L.R. Greenwood (1977).
4. Matzke, M., Proc. 3rd ASTM-Euratom Symposium on Reactor Dosimetry, Ispra, 1.-5.10.1979, p. 721-731 (1979).
5. Shubin, Yu.N., Lunev, V.P., Konobeyev, A.Yu., and Dityuk, A.I., IAEA INDC(CCP)-385, Vienna, Austria (1995).
6. Kim, E.J., Nakamura, T., Uwamino, Y., Nakanishi, N., Imamura, M., Nakao, N., Shibata, S., and Tanaka, S., *J. Nucl. Sci. Techn.*, **36**, 29-40 (1999).
7. Michel, R., Bodemann, R., Busemann, H., Daunke, R., Gloris, M., Lange, H. J., Klug, B., Krins, A., Leya, I., Lüpke, M., Neumann, S., Reinhardt, H., Schnatz-Büttgen, M., Herpers, U., Schiekel, Th., Sudbrock, F., Holmqvist, B., Condé, H., Malmborg, P., Suter, M., Dittrich-Hannen, B., Kubik, P.-W., and Synal, H.-A., and Filges, D., Nucl. Instr. Meth. **B129**, 153-193 (1997).
8. Gloris, M., Michel, R., Sudbrock, F., Herpers, U., Malmborg, P., and Holmqvist, B., Nucl. Phys. **A463**, 593-633 (2001).