

Radioactivity Inventory of the Test Uranium Target at TRIUMF

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Introduction

The main goal of this note is to characterize the radioactivity inventory for the licensing purposes of the low power uranium target at TRIUMF. In this work the Monte Carlo code MCNPX coupled to the material transmutation code CINDER'90, both being products of the LANL (USA), were employed.

This note is organized as follows: Appendix A gives a brief description of the modeling tools and Appendix B provides a number of benchmark examples in order to check the validity of model predictions. Below the target geometry of the test experiment is given together with the proton beam characteristics and irradiation – cooling time periods. Accordingly, in the results section a summarized activation inventory history is provided as a function of different cooling time steps. A particular emphasis is given to the long-lived volatile fission products and heavy residual nuclei being alpha emitters. The very detailed (grouped by atomic masses, elements and individual isotopes) radioactivity inventories are provided in a separate text files, attached to this note.

Target geometry

A cylindrical target geometry was chosen in a rather simplistic way, where 56.71 g of uranium oxide (UO₂: 50.0 g of ²³⁸U and 6.71 g of ¹⁶O with the theoretical density of 10.96 g/cm³) was homogeneously distributed in the “target active volume” of 38.17 cm³. In other words, the active target is 150 mm long with its diameter of 18 mm with its effective uranium oxide density obtained by $56.71\text{g}/38.17\text{cm}^3 = 1.486\text{ g/cm}^3$. This representative production target was centered inside of a metallic ¹⁸¹Ta container with its inner dimensions given by: length of 190 mm and diameter of 18 mm. The wall thicknesses of the container were 0.38 mm from all sides.

Beam and irradiation parameters

The target was irradiated with 500 MeV protons at the nominal beam intensity of 1 μA. The beam profile was taken as Gaussian with its FWHM = 3 mm both in x and y, being a conservative limit of the possible beam focalization, which consequently results in the maximum in-target isotope production rates. The target irradiation history was defined by the “beam on” the target (irradiation) for 300 hours and by the “beam off” the target (cooling) extending up to 100000 hours.

Some results on radioactivity inventory and production yields

Here we present only some representative results while the detailed in-target radioactivity inventory is given in a separate text files both as a function of irradiation time “up” and cooling time “down”.

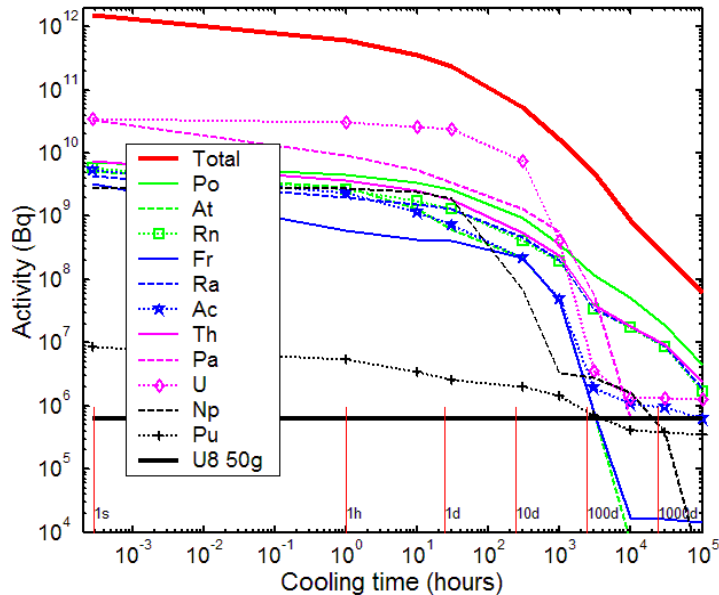


Figure 1: The resulting predicted activity of the test TRIUMF target after 300 h irradiation with 500 MeV protons at 1 μ A. Both total (red solid line) and some important actinide (thin lines) radio-activities are presented as a function of cooling time.

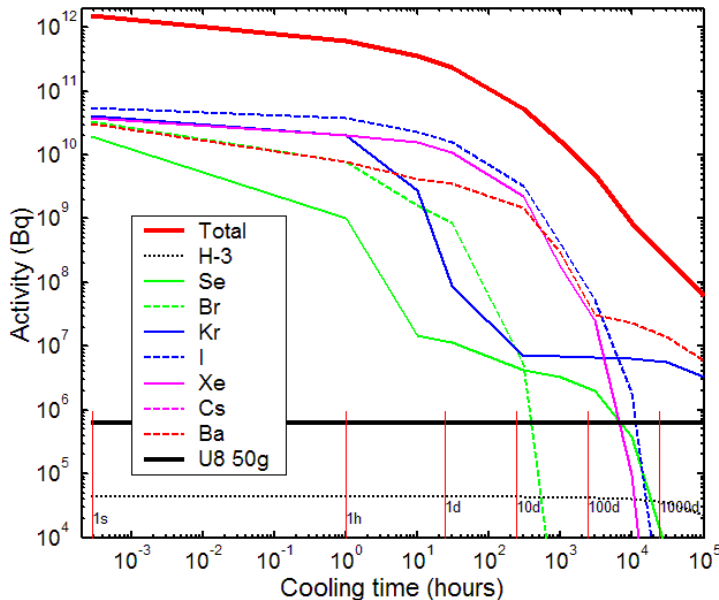


Figure 2: The resulting predicted activity of the test TRIUMF target after 300 h irradiation with 500 MeV protons at 1 μ A. Both total (red solid line) and some important fission product (thin lines) radio-activities are presented as a function of cooling time. The activity due to tritium is presented separately.

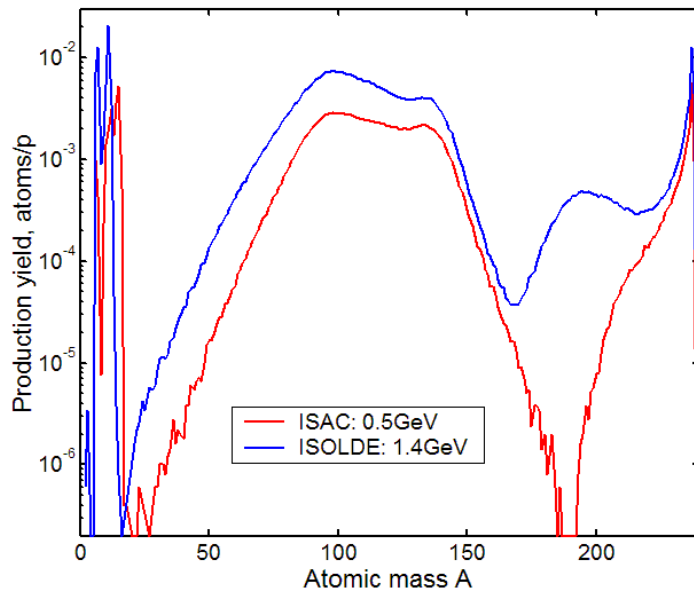


Figure 3: The reaction product mass distributions predicted in the case of the test TRIUMF target (in red; 500MeV protons) together with the reaction product mass distributions predicted in the case of a standard ISOLDE target (in blue; 1400MeV protons). As expected, due to the lower incident proton energy in the case of TRIUMF one observes less fission events (in the mass region from ~40 to ~160) and considerably less heavy residual products (in the mass region from ~190 to ~238).

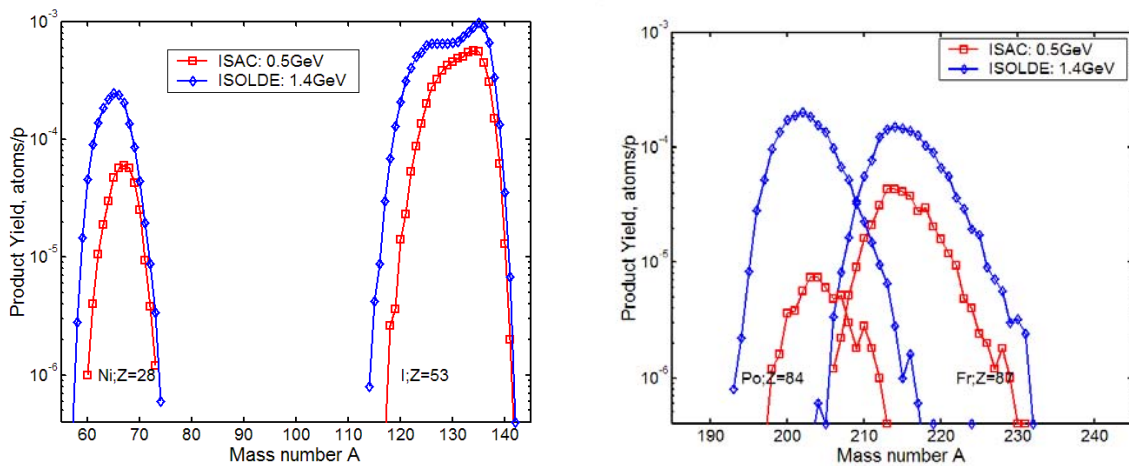


Figure 4: The reaction product isotopic distributions for Ni with I (on the left) and Po with Fr (on the right) predicted in the case of the test TRIUMF target (in red; 500MeV protons) together with the corresponding reaction product isotopic distributions predicted in the case of a standard ISOLDE target (in blue; 1400MeV protons). Higher incident proton energy in the case of ISOLDE results in broader isotopic distributions for fission products (on the left) and suppressed heavy residual nuclei production further from the target nucleus (on the right: note that the decrease in Po production is stronger than in the case of Fr production).

Discussion

Fig. 5 is an illustration of different reactions taking place in the thick target geometries. Indeed, Xe isotopes, being fission products, in the test ISAC target are produced mainly by high energy protons. However, secondary neutrons (both high and low energy) will also contribute to its production as shown explicitly in Fig. 4. As a matter of fact, the total number of fission events in the ISAC test targets is 0.076 fissions/proton, from where low energy neutrons ($E_n < 20\text{MeV}$) are contributing only up to 0.003 fissions/proton. Nevertheless, this small number of fissions is sufficient to dominate the Xe production on the very neutron rich side (see blue line in Fig. 4).

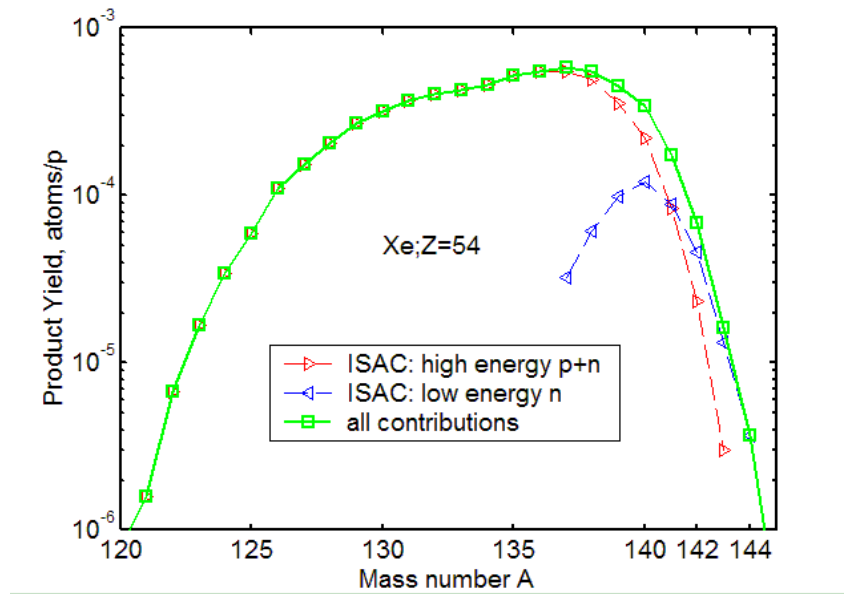


Figure 5: Contribution of different reaction channels to the production of Xe isotopes in the realistic ISAC test target (see the text for details).

This example clearly illustrates the procedure to produce “purified” beams of neutron-rich fission fragments (strongly decreased contamination from stable and neutron deficient Xe isotopes). At ISAC it could be achieved using double-stage target geometry, namely converting protons to neutrons using Ta or W target, while RIB production target (placed next to it) would be exposed only to secondary neutrons. This procedure would also be nearly clean in terms of production of radioactive alpha emitters compared to the direct irradiation of actinide targets with high energy protons. In this particular scenario the geometry of the RIB production target has to be optimized in order to achieve the maximum number of fissions induced by secondary neutrons.

Acknowledgements

The author acknowledges kind hospitality of TRIUMF/ISAC staff during his stay in Vancouver, where this report was prepared.

Appendix A: Calculation tools

In this work the MCNPX (Monte Carlo N-Particle) code [1] has been adopted by the radionuclide inventories and transmutation code CINDER'90 [2], which requires as input a multi-group neutron flux for $E_n < 20\text{MeV}$ and nuclide production rates for reactions at higher neutron energies and for additional particles. In this context MCNPX can yield the medium-energy nucleon-induced spallation and fission products and associated lower-energy neutron flux (see Figure 1 for details).

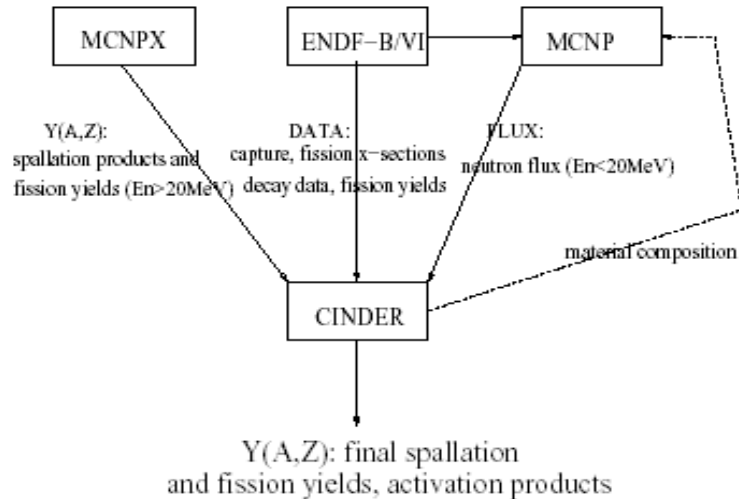


Figure 1: The MCNPX – CINDER code system and its data flow [3].

MCNPX employs the most popular theoretical approach to spallation reactions, namely the intranuclear cascade (INC) and evaporation (EVAP) models. Unfortunately, the transition energy between the INC and EVAP calculation cannot be specified rigorously. For this reason most of the high energy codes, including MCNPX, have looked for an intermediate pre-equilibrium step whose domain of validity is expected to overlap on the INC and EVAP energy domain. Another possibility is to extend the existing data libraries to higher energies from 20 MeV to 150 MeV. This option is also implemented for a number of elements in the recent versions of MCNPX.

The present CINDER'90 code, being the material evolution program, uses a library of transmutation chains based on available nuclear data and evaluations. The library of nuclear data used by CINDER'90, constantly growing in breadth and quality, now describes 3400 nuclides in the range of $0 < Z < 104$. CINDER'90 uses this data: ENDF/B-VI, JEF-1, EAF-3, MDL, GNASH code results, etc.

References

- [1] MCNPX User's Manual, Version 2.5.0, April, 2005, LA-CP-05-0369, LANL, Denise B. Pelowitz, editor also see <http://mcnpx.lanl.gov/>.
- [2] W.B. Wilson and T.R. England, "A Manual for CINDER'90 Version C00D and Associated Codes and Data", LA-UR-00-Draft, April 2001; W.B. Wilson, private communication (2007).
- [3] D. Ridikas, "Optimization of Beam and Target Combinations for Hybrid Reactor Systems and Radioactive Beam Production by Fission", PhD thesis report GANIL-T-99-04 (October 1999).

Appendix B: Benchmark calculations

Neutron production

The neutron production by spallation reaction in different materials has to be well reproduced by simulation codes for both radioprotection and target optimization purposes. It is also an important ingredient for beam intensity calculations both in the single- and in the double-stage production targets. Scattered primary protons and secondary proton production will contribute to the energy deposition and radiation level around the targets and accelerator structures. Equally, it is important to take into account with a good precision the production of other light charged particles as deuterons, tritons, and helium particles being the important contributors to the gas production and damage rates in the target window or other structure materials.

The MCNPX code for particle production was extensively validated within the EURISOL DS project (www.eurisol.org) and we refer the reader to Ref. [1] for more details. Here we present only an example the mostly related to ISAC, namely neutron production from high energy protons interacting with uranium target.

In brief, MCNPX gives reasonable predications of neutron production on uranium target, and this particular observable is little model-dependent.

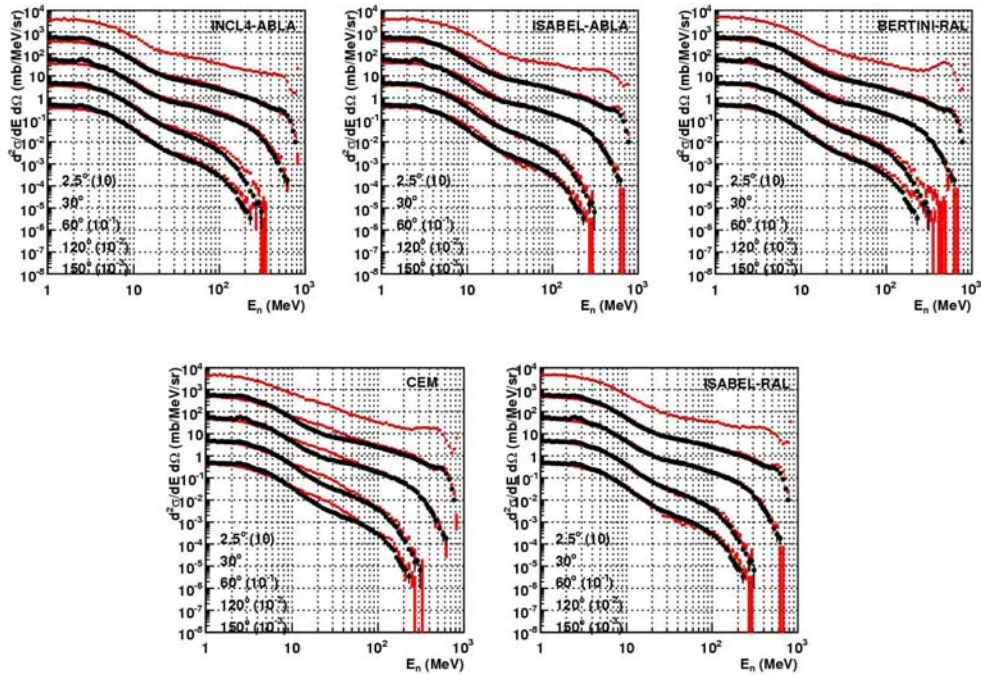


Figure 1 : Neutron double differential cross section in the case of the reaction $U(p,xn)$ at $E_p=800$ MeV. The experimental data are represented in black, and the results of simulation in red. Different model combinations within MCNPX are explicitly given for each figure.

Production of spallation-fission products

Although some valuable experience is already available from presently operating ISOL facilities world wide, the design of a new generation RIB factory requires specific and validated modelling tools. In this context, detailed simulations should be performed in order to optimise the RIB production in terms of target geometry and materials, incident particle types and their

energy, etc. Equally, the radioprotection and safety issues should be addressed and will be based on detailed calculations of particles fluxes, induced radioactivity and resulting dose rates. Two publications [2, 3] within the EURISOL DS project addressed calculations and comparison with experimental data on residue production with the MCNPX2.5.0 transport code, where several spallation-fission models are available. More details on this work can be found in Refs. [2, 3]. Here (in Figs. 1-2) we present only a few most relative examples, which are closely related to ISAC, namely isotope production from high energy protons interacting with uranium targets.

In brief, we conclude that, if the INCL4/ABLA or ISABEL/ABLA physics models are employed within MCNPX, very reasonable predictions of high energy spallation-fission products can be obtained.

References

- [1] B. Rapp et al. “Benchmark calculations on particle production within the EURISOL DS project”, March 8, 2006; report of CEA Saclay, EURISOL DS/Task5/TN-06-04, available at www.eurisol.org.
 [2] J.C. David et al. “Benchmark calculations on residue production within the EURISOL DS project; Part I: thin targets” December 20, 2006; report of CEA Saclay, EURISOL DS/Task11, available at www.eurisol.org.
 [3] J.C. David et al. “Benchmark calculations on residue production within the EURISOL DS project; Part II: thick targets”, June 15, 2007; report of CEA Saclay, EURISOL DS/Task11, available at www.eurisol.org.

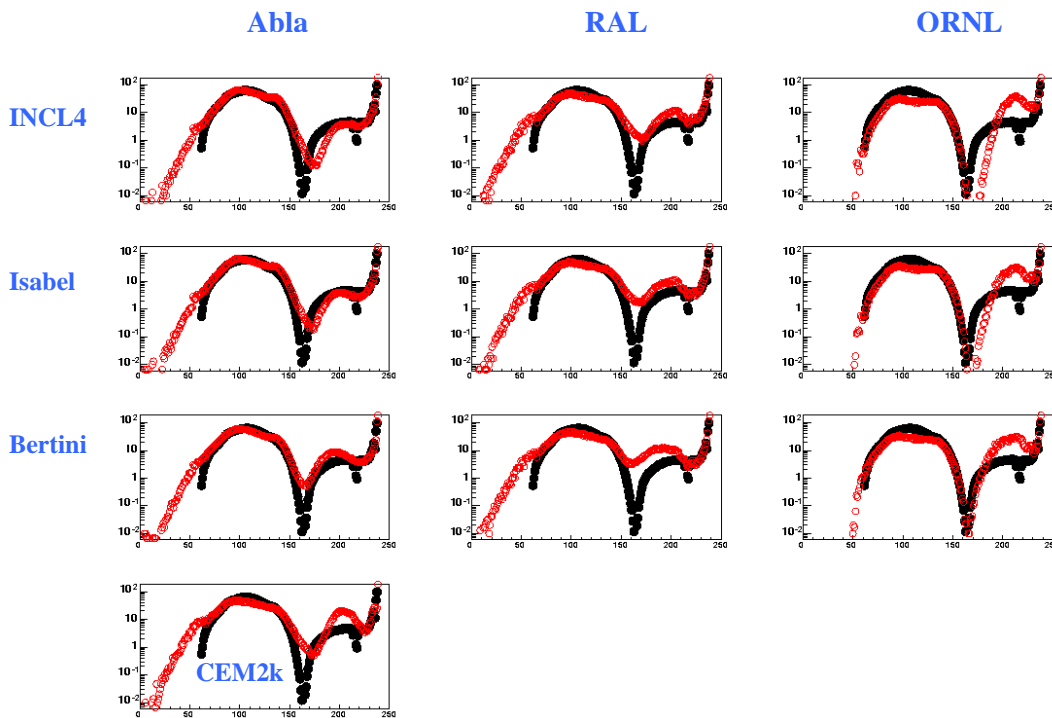


Figure 1: Mass distribution of the reaction products from $^{238}\text{U}(1A.\text{GeV})+p$. GSI Data are in black and model calculations are in red for the ten model combinations available in MCNPX2.5.0. Intra-Nuclear Cascades used are INCL4, Isabel and Bertini (the first three lines), and combined to various evaporation/fission models, which are Abla, Dresner/RAL and Dresner/ORNL (three columns). The last graph (bottom left) is the stand alone results using CEM2k. Cross sections are given in (mb).

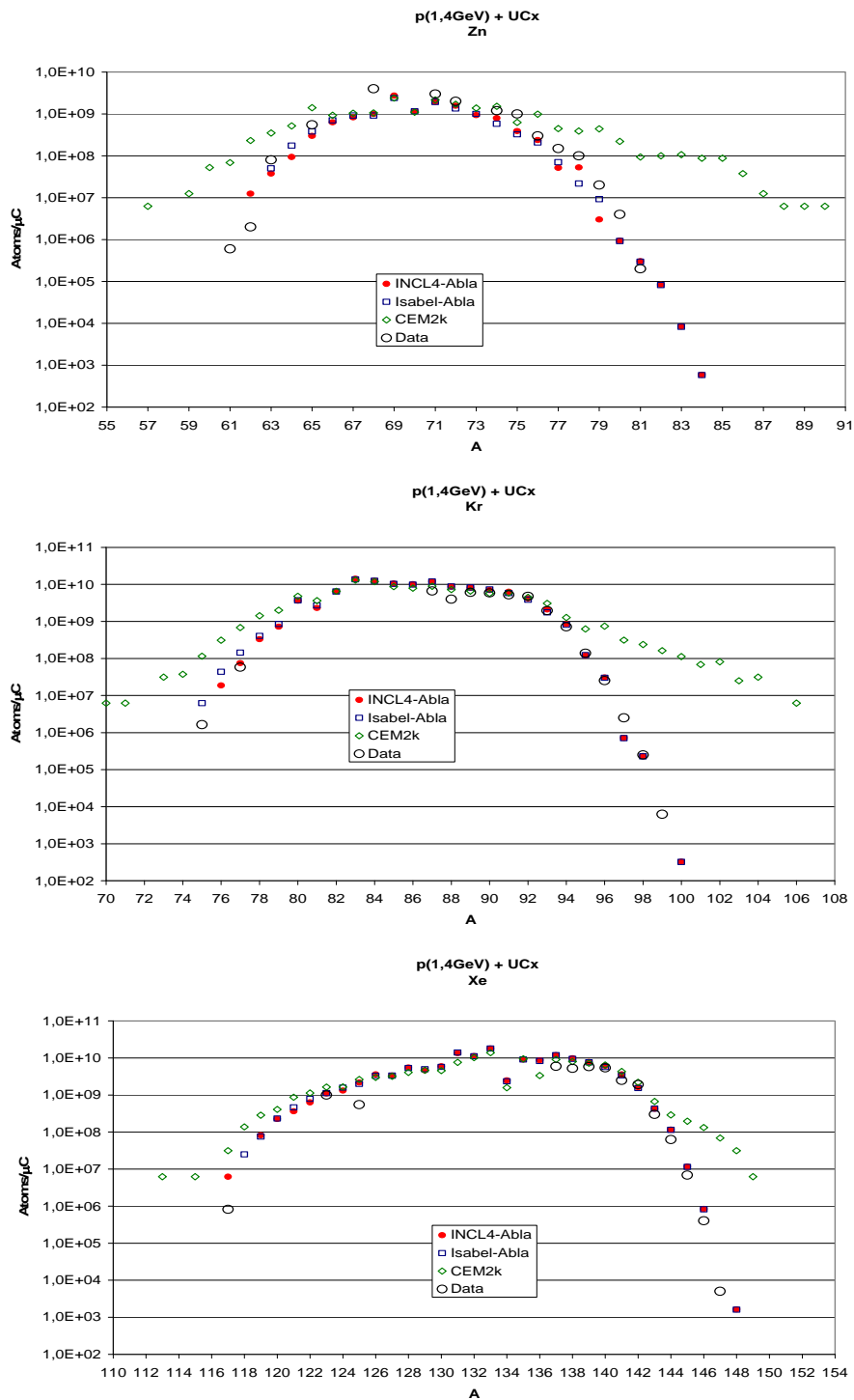


Figure 2: Mass distributions of Zn (top), Kr (middle) and Xe (bottom) obtained with the UC_x thick target impinged by 1.4 GeV proton beam. Data (black open circles) are from ISOLDE and calculations were made with MCNPX2.5.0. Three different model combinations were examined: INCL4-Abla (red full circle), Isabel-Abla (blue open square) and CEM2k (green open diamond).