Abstract

The GEANT4 code offers an extensive set of hadronic models for various projectiles and energy ranges. These models include theoretical, parameterized and, for low energy neutrons, data driven models. Theoretical or semi-empirical models sometimes cannot reproduce experimental data at low energies (<100MeV), especially for low Z elements, and therefore recent GEANT4 developments included a new particle hp package which uses evaluated nuclear databases for proton interactions below 200MeV. These recent developments have been used to study target designs for low energy proton accelerators, as replacements of research reactors, for medical applications. Presented in this paper are results of benchmarking of these new models for a range of targets, from lithium neutron production targets to molybdenum isotope production targets, with experimental data.

INTRODUCTION

Currently the production of medical tracer isotopes for use in imaging techniques such as SPECT (Single Photon Emission Computed Tomography) and PET (Positron Emission Tomography) [1] relies principally upon an aging fleet of nuclear reactors. For example the most common medical isotope, $^{99m}$Tc, used in over 80% of all radiopharmaceutical procedures, is currently produced, via its generator $^{99}$Mo, by nuclear research reactors such as NRU-Canada and HFR-The Netherlands, which together produce over 60% of the world’s $^{99m}$Mo/$^{99}$Tc supply. Both of these reactors are old (>50yrs) and close to decommissioning, but as yet there is no real replacement in place [2] [3]. There is considerable concern that we will soon be facing a similar situation to that of the 2010 isotope crisis, when both reactors were offline simultaneously resulting in a significant decrease in the supply of $^{99m}$Tc and the postponing or cancelation of many vital radioisotope procedures [1] [2] [3] [4] [5].

We believe that the solution to this impending problem could lie in accelerator-based production methods, of both $^{99m}$Tc and possible replacement isotopes. A collaboration with Siemens is focusing on the potential of a compact, low energy proton device for the generation of radioisotopes [6]. A study of optimal target designs for such a system has been undertaken using GEANT4 simulations of low energy (<10MeV) proton induced reactions.

GEANT4

GEANT4 is a well-known, well-used toolkit for the simulation of particle interactions in numerous areas of physics, especially in its origins in high-energy physics. However there has been very little implementation of GEANT4 in simulating the interactions of low energy protons with targets. Moreover, the standard physics models available with the current release of GEANT4 for these types of simulation, such as the QGSP_BIC_HP and QGSP_BERT_HP, are all theoretical [7] [8] [9] [10]. Initial studies of low energy proton interactions with thick (0.7mm) targets comprised of lithium or beryllium have shown that these theoretical models breakdown in the low energy limits. In light of this, a new data driven model, QGSP_BIC_PHP, has been developed to simulate the interactions of protons with energies less than 100MeV with targets. The data for the new model is derived from either the ENDF or TENDL libraries, and is selected by the environmental settings for each simulation.

The initial light element target/neutron production benchmarking was repeated for the new model before a second phase of benchmarking for heavier element targets was undertaken. In this second phase thin foil targets were used for the heavy element targets with proton beams of energy less than 10MeV. In one case the experimental cross sections were so low that a higher energy (<100MeV) proton beam was necessary and hence a thicker pellet type target was used.

This paper presents the results of benchmarking of the new data driven model for a range of targets, including the initial light element targets for neutron production and for several medical radioisotopes produced using accelerator based methods for which reasonable experimental data is available for comparison.

RESULTS

Neutron Production

Initial benchmarking results for both theoretical and data driven models can be seen in fig.1, overlaid with experimental data [11] [12] [13]. It can be seen how the theoretical model completely breaks down for light target low energy proton interactions, significantly underestimating the neutron production from thick lithium and beryllium targets by approximately 3 orders of magnitude. However the data driven model almost perfectly replicates the experimental data for both targets.

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Isotope Production

In the case of radioisotope production a more intensive benchmarking of these models was carried out for these using a range of heavier targets. The simulation and experimental results shown below, Figs.2-5, are for several isotopes currently of interest for medical applications. These isotopes are currently being produced using accelerator-based methods or such production methods are being investigated.

The lightest isotope to be tested is $^{64}\text{Cu}$, a diagnostic PET isotope used to distinguish malignant tumour tissue [2] [14], which can be produced through the $^{64}\text{Ni}(p,n)^{64}\text{Cu}$ reaction. The results are shown in fig.2. It can be seen that while there is good agreement between the experimental [15] [14], theoretical and data driven cross sections at the higher energy range (7-10MeV), the theoretical models start to break down as the energy decreases (<5MeV) at which no isotope production is seen. The data driven model continues to show good agreement with the experimental data down to approximately 3MeV where the cross section becomes too low (<1mb) to obtain simulated production.

Next, the production of $^{89}\text{Zr}$, a PET diagnostic isotope often used in connection with labelled antibodies [16], from the $^{89}\text{Y}(p,n)^{89}\text{Zr}$ reaction was simulated. The results can be seen in fig.3. Again, at the higher energies (7-10MeV) there is reasonable agreement between both simulation results and the experimental data [17] [14]. However the theoretical model starts to break down at approximately 6MeV. The data driven model still shows isotope production down to 4MeV for this reaction.

Possibly the most important test case is the simulation of the production of the $^{99m}\text{Tc}$ generator via the $^{100}\text{Mo}(p,pn)^{99}\text{Mo}$ reaction. A range of experimental data is available from the literature [17] [18] [19] and online libraries (such as EXFOR), some examples of which can be seen in fig.4. This example shows how the available data sources can affect the simulation results obtained using the
data driven model and with such a range of experimental results from which to choose a sensible comparison between experimental and simulated data is not always possible. In such cases the theoretical model may provide more reliable results.

![Figure 5: Experimental and simulation results for the $^{123}$Te(p,n)$^{123}$I reaction.](image)

The final test case in this phase was the reaction $^{123}$Te(p,n)$^{123}$I, producing an isotope currently being used for thyroid SPECT imaging [2]. It can be seen from the results in fig.5 that both simulation models overestimate the experimental [20] isotope production cross-sections in the less than 10MeV energy region, the theoretical results more so than the data driven model.

**CONCLUSION**

In this paper we have introduced a data driven model for the simulation of accelerator driven radioisotope production at low proton energies. The benchmarking results presented are encouraging and indicate that a successful low energy data driven model can indeed be used successfully in GEANT4 to simulate low energy proton interactions. However the test cases presented here are limited and a much more stringent validation process must yet be carried out before this new model can be included in the standard GEANT4 release with confidence.

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**REFERENCES**


