DESIGN AND TEST OF AN ACCELERATOR DRIVEN NEUTRON ACTIVATOR AT THE JRC CYCLOTRON OF THE EUROPEAN COMMISSION

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Abstract

A compact, accelerator-driven, neutron activator based on the Adiabatic Resonance Crossing (ARC) method has been developed in the frame of the INBARCA project, funded under the EUREKA programme. It has been assembled and tested at the Scanditronix MC40 Cyclotron of the Joint Research Centre (JRC Ispra, Italy) with the aim of efficiently utilising ion-beam generated neutrons for the production of radioactive nanoparticles for brachytherapy. Monte Carlo and Computational Fluid Dynamics simulations were carried out to optimise the design of the activator, which is based on a hybrid approach, coupling a lead buffer and a graphite reflector. The activator has been tested under various experimental conditions, and the neutron fluxes at the different irradiation positions of the system were calculated. Experimental results on the activation of pure Au, Al, Mo, Ho and Re foils are presented and show a good agreement with calculations. The results obtained are compared to previously reported work. The next experimental stage will be the activation of Ho/Re Poly-Lactic Acid nanoparticles for brachytherapy applications.

INTRODUCTION

Proton-beam driven neutron sources are the main component of Accelerator Driven Systems [1] and Spallation Neutron Sources [2]. The TARC (Transmutation by Adiabatic Cross Resonance) concept was introduced by C. Rubbia and has been experimentally verified by using a 3.5 GeV proton beam and 334 tons of high-purity lead [3-5]. The TARC experiment was seen as a milestone in the development of the minor actinides transmutator, but it also opened new perspectives for the design of an accelerator driven activator to be used for the production of radioisotopes for cancer therapy, that are more typically produced in nuclear reactors. Recently, an ARC activator prototype designed for medical radioisotopes production has been tested using a 65 MeV proton beam [6].

In the frame of the INBARCA (Innovative Nanosphere Brachytherapy using Adiabatic Resonance Crossing with Accelerators) project [7] a modified ARC prototype has been developed to validate concepts, components and design tools in order to explore the feasibility of activation of Re (^{186g}Re, $T_{1/2}$ = 89.25h, ^{188g}Re, $T_{1/2}$ = 16.98h) and Ho (^{166}Ho, $T_{1/2}$ = 26.80h) radioisotopes embedded in Poly-Lactic Acid micro- or nano- particles suitable for cancer brachytherapy [8]. The activator prototype has been installed on a dedicated beamline of the Scanditronix MC40 Cyclotron of the Institute for Health and Consumer Protection (IHCP) at JRC Ispra, the nominal beam characteristics of which are given in Table 1. The preliminary tests show satisfactory agreement with the Monte Carlo simulations. The main results were also compared to those obtained in the previously reported ARC experiment [6].

Table 1: Beam characteristics of the JRC Cyclotron

Particle	Energy (min.-max.) (MeV)	Maximum Extracted Current (μA)
p	8 – 40	60
d	8 - 40	30
$^{4}He^{2+}$	8 - 53	30
$^{3}He^{2+}$	4 - 20	60

DESIGN AND INSTALLATION OF THE NEUTRON ACTIVATOR FACILTY

MCNPX [9] and FLUKA [10-11] Monte Carlo (MC) codes were used to simulate the main components (target, water cooling mass, lead buffer and graphite reflector) of the activator, in order to obtain the maximum yields for the radioisotopes of interest. The combined neutron capture in both epithermal and thermal energy ranges was optimised by using, besides the ARC effect, a properly dimensioned graphite neutron reflector. A set of coupled Monte Carlo/CFD (FLUKA and Star-CD) simulations have been carried out in order to properly dimension the water cooling systems with respect to heat removal and neutron moderation properties. As reported in the conceptual drawing of Fig. 1, a fast neutron flux is first generated in a Be target by the proton beam, then it is down scattered, with relatively low parasitic capture, using the assembly of lead and graphite surrounding the Be target. The material to be activated is located in the irradiation channels in selected positions where the

neutron flux has the optimum characteristics for the neutron capture reactions of interest.

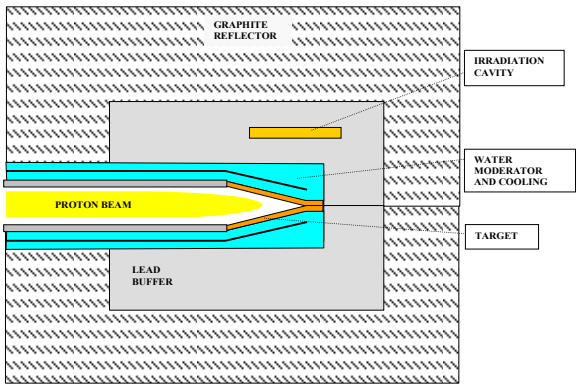

Figure 1: Schematic drawing showing the main components of the neutron activator facility

A reference simulation of the complete activator with a proton beam of 36 MeV estimates the average neutron flux in the lead buffer volume at 5.07 x 10^8 n.cm^{-2}.s^{-1} μA^{-1} (1.77 x 10^{10} n.cm^{-2}.s^{-1} with 35 μA beam current), whereas the neutron flux in an irradiation channel has been estimated at 1.26 x 10^9 n.cm^{-2}.s^{-1} μA^{-1} (4.42 x 10^{10} n.cm^{-2}.s^{-1} with 35 μA).

The neutron activator prototype was constructed and installed on a dedicated cyclotron beamline, as shown in Fig. 2. It is equipped with an electronic control system to monitor beam current, beam shape and position, target temperature and water cooling parameters. In the case of abnormal conditions an interlock signal switches off of the cyclotron ion source. As can be seen in Fig. 2, eight irradiation channels (diameter of 10 mm) are located in the buffer to host the samples for activation, which are inserted by means of Al-clad graphite pins.

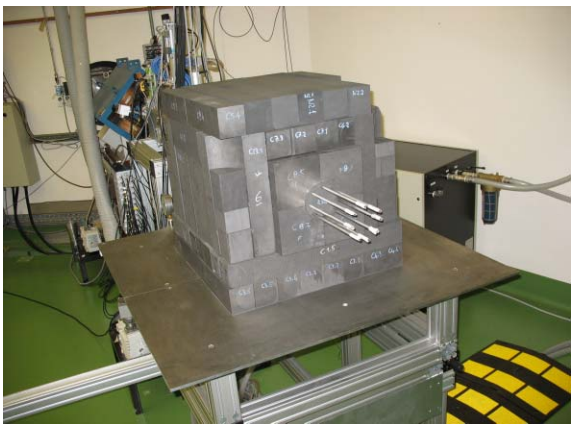

Figure 2: The assembled activator showing the eight pins used to insert samples into the activation channels

PRELIMINARY TESTS AND RESULTS

The system was progressively tested with several runs using (1) bare target only, (2) water cooled target (target + moderator), and (3) complete configuration, in order to test the various sensors and interlocks. In addition, for each run Au metallic foils were employed as neutron flux monitors in order to monitor the effect of each component on the neutron flux and energy distribution. It was observed that the relative yield of the ^{197}Au(n,γ)^{198g}Au reaction to the ^{197}Au(n,2n)^{196g}Au reaction increased dramatically as the moderator and reflector components were added, as expected. The results of these measurements will be fully analysed and compared to theoretical calculations in a future paper.

Activation yields were measured by γ-ray spectrometry using calibrated Ge detectors. The yields for Au, Al, Mo, Ho and Re metallic foils, irradiated in the fully constructed system, are reported in Table 2 along with some comparative values from reference [6].

Table 2: Comparisons of the activation yields of Au, Al, Mo, Ho and Re foils (in kBq.g^{-1}.μA^{-1}.h^{-1}). The uncertainty in measured yield was less than 10%.

Reactions	MCNPX yield	Exp. Yield (Ispra)	Exp. yield (LLN)
^{197}Au(n,γ)^{198}Au	553.83		
^{197}Au(n,γ)^{198g}Au		724.20	194.46 * (526.99 **)
^{197}Au(n,2n)^{196}Au	1.99		
^{197}Au(n,2n)^{196g}Au		1.58	
^{27}Al(n,α)^{24g}Na		5.37	
$^{98/100}$Mo(n,γ/2n)^{99}Mo		7.13	1.85 * (5.02 **)
^{165}Ho(n, γ)^{166}Ho	1650.00		
^{165}Ho(n, γ)^{166g}Ho		2520.00	
^{185}Re(n, γ)^{186}Re	693.00		
^{185}Re(n, γ)^{186g}Re		385.00	
^{187}Re(n, γ)^{188}Re	1150.00		
^{187}Re(n, γ)^{188g}Re		2100.00	

** Yield corrected for proton energy according to [12]*

*** Yield for 65 MeV proton energy [6]*

Since MCNPX does not consider the isomeric states, the calculated yields are the sum of the yields of all the isometric states while the experimental reported ones are specific to a given state. Although the two activator facilities are different, it can be noted that the activation yields for ^{198g}Au and ^{99}Mo (generator of ^{99m}Tc) radioisotopes obtained in this work are almost 4 times higher that those obtained at LLN, if one includes the neutron yield correction due to the proton beam energy [12] (65 MeV used at LLN compared to 36 MeV at Ispra). A preliminary calculation on the unfolded neutron energy distribution from the foil activation yields estimates a neutron flux of 4.93 x 10^9 n.cm^{-2}.s^{-1}.μA^{-1} in the irradiation cavity, which is in is good agreement with the simulated flux value.

CONCLUSION AND PERSPECTIVES

A new design concept of an accelerator driven neutron activator has been constructed and tested. Preliminary results on activations of foils of various materials were found to be in good agreement with the theory.

The promising activation yields accomplished in this work open interesting perspectives for SPECT imaging and brachytherapy studies using Ho and Re based radioactive nanoparticles. When considering reported work on the testing of the ARC concept, this work constitutes an important step toward its use for production of medical radioisotopes. With respect to other facilities made of several cubic metres of lead, the neutron activator developed in this work is quite compact (60 x 60 x 60 cm^3, weight ~ 600 kg). Furthermore, with reference to the literature, higher production yields of ^{99}Mo (generator of ^{99m}Tc) and ^{198g}Au were obtained.

Further irradiation tests are being carried out to investigate ways of increasing radioisotope yields by, for example, changing the cyclotron beam (e.g. deuterons), or considering other target material for neutron generation (e.g. Ta). While nuclear reactors clearly out-perform cyclotron-driven ARC systems regarding radioisotope production yields, the neutron activator developed in this work constitutes an interesting alternative for production of medical radioisotopes, and in combination with high-current cyclotrons could be of considerable advantage in particular cases.

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