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Production of ^{28}Mg by bombardment of ^{nat}Cl with 200 MeV protons: Proof-of-concept study for a stacked LiCl target

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HIGHLIGHTS

- Radionuclidically pure Mg-28.
- LiCl stacked targets irradiated in a 200 MeV proton beam.
- Monte Carlo modelling of particle fluence.
- Removal of Na contaminants from final Mg-28 product.

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ABSTRACT

A stacked target consisting of ten Al-encapsulated LiCl discs, for producing ^{28}Mg via the ^{nat}Cl(p,X)^{28}Mg process in the energy region 50–200 MeV, is described. This target was irradiated with a 200 MeV beam at an intensity of 100 nA, providing information on both yield and outscattering losses. Results of a Monte Carlo modelling of the beam and target, by means of the code MCNPX, are also presented. Similar Al-encapsulated LiCl discs were individually irradiated with 66 MeV proton beams of 65 and 90 μA, respectively, to study their behaviour under high-intensity bombardment. Once removed from the Al encapsulation, the ^{28}Mg can be separated from the LiCl target material efficiently, using a 12.5 cm x 1 cm^2 column containing Purolite S950 chelating resin. The eluate contains ^{7}Be but no other measurable radiocontaminants. The removal of the ^{7}Be contaminant is performed by cation exchange chromatography in malate media, with ^{28}Mg being retained by the resin and ^{7}Be eluted.

1. Introduction

The radionuclide ^{28}Mg has been employed as a radiotracer of magnesium since the second half of the previous century (Sheline and Johnson, 1953) and found wide application in the study of absorption, retention and excretion in metabolic processes (cf. Martin and Bauer, 1962; Mendelson et al., 1965; Kniffen et al., 1972; Watson et al., 1979; Verhas et al., 2002) as well as in animal and human physiology (cf. Schimansky, 1973; Schwartz et al., 1981; Iwata et al., 1992; Heijnen et al., 1996). Magnesium is an essential nutrient for all living organisms. Since ^{28}Mg ($T_{1/2}$ = 20.9 h) is its only radionuclide with a half-life longer than 10 min, it has regularly been employed in quantitative studies of Mg bioavailability in agriculture and nutrition (cf. Verhas et al., 2002; Bohn, 2003, and references therein). Its mode of decay is 100% by β^- emission (Firestone and Eckström, 2004) with several intense γ-lines suitable for γ-ray spectrometry.

The present investigation concerns the production of ^{28}Mg with a cyclotron up to a kinetic energy reach of k = 200 MeV. Probst et al. (1976) provided a summary of the various possible production routes, which is still valid today. Two of these found application in the routine production of no-carrier-added ^{28}Mg, namely ^{nat}Cl(p,X)^{24}Mg and ^{27}Al(α,3p)^{24}Mg. The latter reaction is very convenient due to the use of uncomplicated Al metal targets, while the former process typically requires the use of compressed and encapsulated chloride salt targets. As the magnetic rigidities of protons and α-particles are the same, one can compare the relevant excitation functions and integral yields directly. Such a comparison is presented in this work with the aim to determine in which energy window the α-particle route has the advantage and at which energies the proton route will provide higher yields.

In the case of protons, the work of Lundqvist and Malmborg (1979) deserves a special mention. These authors investigated the production of carrier-free ^{28}Mg and ^{24}Na in proton-induced

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reactions on natural targets of Si, P, S, Cl, Ar and K in the energy region 50–180 MeV. It was shown that only the ^{nat}P(p,X)^{28}Mg process has higher cross sections for ^{28}Mg formation than ^{nat}Cl(p,X)^{28}Mg (about 30% higher at 150 MeV). Unfortunately, many compounds of phosphorus decompose at relatively low temperatures, rendering them unsafe to use as target materials. In fact, we could not identify a single suitable compound with a relatively high P content as well as good thermal stability. The conclusion was that ^{nat}Cl + p was the production route of choice and LiCl was the target material of choice based on thermal stability and Cl content (Steyn et al., 2008).

LiCl has excellent thermal properties for targetry, including a melting point of 605 °C and a boiling point of 1325 °C. A yield of about 80% of the ^{nat}Cl + p theoretical maximum can also be expected. The problem with a large energy window, such as 50 – 200 MeV, is that a single LiCl target would be about 148 mm (30.6 g/cm^2) thick. Such a thick target would be impractical and impossible to cool efficiently during a high-intensity bombardment. A solution was suggested in our previous paper (Steyn et al., 2008), namely to stack several thinner, encapsulated LiCl targets behind one another and to provide each individual target with fast flowing cooling water in a 4π geometry. In this way, the total surface area to cool from can be increased significantly, albeit with the loss of some production yield due to the capsule walls and cooling water "dead layers". This concept is, of course, nothing new – the Brookhaven Linac Isotope Producer (BLIP), for example, has bombarded stacks of multiple targets since 1973 (Mausner et al., 1984, 1990). The BLIP targets are relatively large, however. Mausner et al. (1990) reported compressed salt discs of 4.5 and 7 cm in diameter, enclosed in stainless steel capsules with Inconel windows. Compressed KCl and RbCl targets were developed for the production of ^{28}Mg and ^{82}Sr, respectively and it was reported that the BLIP was the only supplier of ^{28}Mg in North America at that time.

In order to reduce the amount of irradiated target material that needed to be processed, it was decided to limit the diameter of individual target discs to 20 mm, albeit with some beam losses towards the back of the stack due to the radial beam spread caused by multiple Coulomb scattering inside the target material. The philosophy is that some outscattering of protons can be tolerated, as long as these losses are not too severe. Another reason why target discs of 20 mm in diameter were selected was because such targetry would be compatible with the existing infrastructure for target handling, bombardment and batch processing in the production of short-lived radionuclides as well as with existing methods for encapsulation and decapsulation in our facility at iThemba LABS. We report here on an experimental target holder designed to irradiate a stack of encapsulated LiCl targets with a 200 MeV proton beam. In addition, the individual targets were compatible with the standard production target holders in use on the horizontal-beam target station (Target Station 1) for bombardments with 66 MeV protons (Steyn et al., 2013).

The work reported here on the ^{nat}Cl(p,X)^{28}Mg production route had three clearly defined objectives: First, to determine the outscattering losses of a 200 MeV proton beam experimentally in an actual target stack, as well as by means of Monte Carlo modelling using the radiation transport code MCNPX (Pelowitz, 2011). Second, to investigate individual target performance in bombardments with moderately intense beams, employing a 66 MeV proton beam. Third, to develop a chemical separation technique using the activated target material obtained from these bombardments. This proof-of-concept study would assist in deciding on a production regimen. In addition, production yields would be obtained which could be compared with expectations based on the already completed nuclear data study (Steyn et al., 2008). The choice of 66 and 200 MeV beam energies was influenced by the capabilities of the present cyclotron infrastructure as well as with the weekly beam schedule of the k=200 separated sector cyclotron (SSC) at iThemba LABS. The 200 MeV beams are regularly produced for proton therapy. The 66 MeV beams are used for the routine production of radionuclides and for neutron therapy. Some of the ancillary infrastructure necessary to produce beams of high intensity (e.g. flat-top RF resonators, RF bunchers, etc. – see Steyn et al., 2013) had been designed and implemented for 66 MeV beams only and not for any other proton energies at this stage. Thus, the SSC cannot produce 200 MeV beams of high intensity without further development. The current work should therefore be seen in the light of a proof-of-concept study. Production yields achievable with the SSC at its current level of evolution, for different target and beam configurations, are also reported.

The relevant excitation function guided the choice of the target configurations chosen for this work, as discussed in Section 2. The experimental investigations at 200 MeV and 66 MeV are discussed in Sections 3 and 4, respectively. The method employed for the radiochemical separation of ^{28}Mg from the bombarded LiCl targets is discussed in Section 5. A summary and conclusion is presented in Section 6.

2. Nuclear data and target configurations

Only two datasets exist in the EXFOR library (IAEA, 2015) on the excitation function for the ^{nat}Cl(p,X)^{28}Mg process, by Lundqvist and Malmborg (1979) and by Steyn et al. (2008). The earlier study contains the only thin-target point data ever to be reported for this process, to our knowledge, while the latter investigation derived the excitation function by means of a numerical differentiation of an experimental integral yield curve. In Fig. 1, these data are compared with theoretical predictions as given in the TENDL-2015 compilation, based on calculations with the TALYS code (Koning et al., 2015). The theoretical curve underpredicts the experimental data by almost a factor of 5 but has approximately the right shape. (Note that the theoretical curve has been multiplied by a factor of 5 for purposes of comparison with the experimental data.) The excitation function rises monotonically from threshold and flattens out towards higher energies, with no

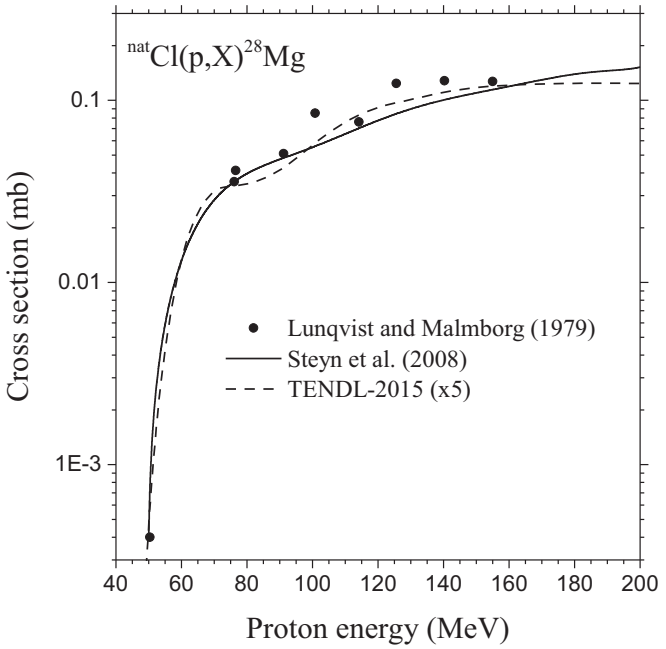

Fig. 1. Excitation function for the production of ^{28}Mg in the bombardment of ^{nat}Cl with protons. The TENDL-2015 values have been multiplied by a factor of 5. The solid curve has been used to calculate the integral yield (see text).