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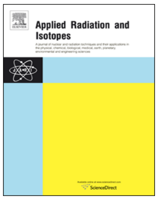

Nuclear model analysis of excitation functions of proton, deuteron and α-particle induced reactions on nickel isotopes for production of the medically interesting copper-61

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HIGHLIGHTS

- Evaluation of ^{61}Ni(p,n)^{61}Cu, ^{62}Ni(p,2n)^{61}Cu, ^{60}Ni(d,n)^{61}Cu and ^{58}Ni(α,p)^{61}Cu reactions.
- Nuclear model calculations (TALYS and EMPIRE) and fitting of excitation functions.
- Estimation of integral yield and impurity level in the production of ^{61}Cu.
- Validation of the evaluated data of (p,xn) reactions by comparison with the measurements on ^{nat}Ni.

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ABSTRACT

Excitation functions of the ^{61}Ni(p,n)^{61}Cu, ^{62}Ni(p,2n)^{61}Cu, ^{60}Ni(d,n)^{61}Cu and ^{58}Ni(α,p)^{61}Cu reactions were analyzed with respect to the production of ^{61}Cu ($T_{1/2}$=3.33 h), a promising radionuclide for PET imaging. The nuclear model codes EMPIRE and TALYS reproduced the experimental data of all reactions well, except those for the (d,n) process. The fitted excitation functions were employed to calculate the integral yield of ^{61}Cu in all reactions. The amounts of the possible impurities ^{62}Cu and ^{60}Cu were assessed. A validation of the evaluated (p,xn) data was attempted.

1. Introduction

The significance of positron emission tomography (PET) in diagnostic nuclear medicine has considerably increased in recent years. This non-invasive procedure is generally performed using pharmaceuticals labeled with short-lived positron emitters of light elements that are usually parts of organic compounds [i.e. ^{11}C ($T_{1/2}$=20 min), ^{13}N ($T_{1/2}$=10 min), ^{15}O ($T_{1/2}$=2 min) and ^{18}F ($T_{1/2}$=110 min)]. A limitation with regard to widespread growth of PET applications is the short half-lives of the above mentioned radionuclides. Except for ^{18}F, for production of the other radionuclides an on-site cyclotron is essential (cf. Qaim, 2003, 2004). Recently, there has been a growing interest in the development of longer lived positron emitters (termed as *non-standard positron emitters*). Applications of those radionuclides extend from the study of slow metabolic processes to labeling of organic compounds leading to analog tracers (e.g. with halogens) and quantification of targeted therapy (Qaim, 2011, 2012). The decay characteristics of those non-standard positron emitters, including positron energy, the positron decay fraction, prompt emission of associated gamma rays and longer half-lives, determine the qualitative and quantitative accuracy (i.e. blurring, spatial resolution, sensitivity, radiation dose, etc.) of the image, which ultimately defines their possible utilization in PET (Laforest et al., 2002). However, drawbacks associated with some of their decay characteristics can be suppressed or overcome significantly by the development of efficient image reconstruction algorithms and permitting an extended imaging period (Herzog et al., 2006, 2008; Ruangma et al., 2006; Laforest and Liu, 2008).

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Several copper radioisotopes (including ^{60}Cu ($T_{1/2}$=23.7 min; $I_{\beta+}$=92%; $E_{\beta+}$=2.940 MeV; EC=8%); ^{62}Cu ($T_{1/2}$=9.7 min; $I_{\beta+}$=93%; $E_{\beta+}$=2.925 MeV; EC=7%); ^{64}Cu ($T_{1/2}$=12.7 h; $I_{\beta+}$=17.8%; $E_{\beta+}$=0.657 MeV; EC=43.8%; I_{β^-}=38.4%) and ^{67}Cu ($T_{1/2}$=61.8 h; I_{β^-}=100%)) find extensive medical applications (Blower et al., 1996; Anderson et al., 2003; Rowshanfarzad et al., 2006). ^{61}Cu ($T_{1/2}$=3.33 h) is another radionuclide of copper with potential utilization in nuclear medicine (Szelecsényi et al., 1993; McCarthy et al., 1999; Rowshanfarzad et al., 2006; Thieme et al., 2013). It decays mainly via positron emission ($I_{\beta+}$=62%; $E_{\beta+}$=1.159 MeV; EC=38%) with accompanying dominant γ-rays of 283 keV (12%) and 656 keV (10.8%). It is considered to be a suitable candidate for studies of slow kinetics of larger proteins, such as peptides and antibodies, or cells (McCarthy et al., 1999; Williams et al., 2005). ^{61}Cu could yield higher quality images than with ^{64}Cu, due to the higher positron decay branching and the shorter half-life. The radiation dose associated with ^{61}Cu is comparable with that of ^{18}FDG (Williams et al., 2005). The associated disadvantage with ^{61}Cu is the high end point energy of the positron and prompt γ-rays which result in the degradation of the spatial resolution and increased noise of the PET image. However, these deficiencies can be reduced by using the recently developed novel image reconstruction algorithms (e.g. Williams et al., 2005; Ruangma et al., 2006; Herzog et al., 2008; Laforest and Liu, 2008).

Several charged-particle induced reactions on enriched nickel and zinc isotopes have been demonstrated to produce pure no-carrier-added ^{61}Cu. Targets of natural Ni, Zn and Co have also been used at various cyclotrons to produce this radionuclide (Szelecsényi et al., 2005; Rowshanfarzad et al., 2006). In this work proton, deuteron and alpha-particle induced reactions on enriched nickel targets have been considered. The four investigated reactions include ^{61}Ni(p,n)^{61}Cu, ^{62}Ni(p,2n)^{61}Cu, ^{60}Ni(d,n)^{61}Cu and ^{58}Ni (α,p)^{61}Cu. For each reaction a critical evaluation of the excitation function was done. As *a priori* the general behavior of the investigated reaction was checked by comparing the measurement with the results of the TENDL file (Koning et al., 2012) and the nuclear model code ALICE-IPPE (Dityuk et al., 1998). The experimental data were also compared with the results of nuclear model codes, TALYS and EMPIRE, with nuclear model parameters adjusted within their recommended limits to improve the agreement with experimental data. Therefrom recommended reaction crosssection values for the production of ^{61}Cu were deduced via statistical fitting of the selected data. Those data were used to derive the integral yield of the desired radionuclide. Nuclear model calculations for some of the above mentioned reactions were also performed by Sadeghi et al. (2012), but those results were mainly for natural targets and not detailed. The reactions leading to the formation of significant radionuclidic impurities, ^{60}Cu and ^{62}Cu, were also analyzed in this work. Those short-lived radionuclides also find application in diagnostic imaging. Furthermore, the suggested excitation functions for the proton induced reactions were validated by comparison with the experimental cross sections from natural targets.

2. Nuclear model calculations and statistical fitting

Theoretical calculations of the cross sections were performed by two nuclear model codes, namely EMPIRE 3 (Herman et al., 2007) and TALYS 1.4 (Koning et al., 2008). In both the codes the nuclear structure, optical model, discrete levels and deformation parameters were mainly retrieved from the Reference Input Parameter Library (RIPL-3) of the IAEA (Capote et al., 2009).

The nuclear reaction code system, EMPIRE 3 contains several codes to describe the important nuclear reaction mechanisms. In all the reaction calculations discrete level schemes were adjusted by using the FITLEV option. The option of EMPIRE-global specific model which is based on enhanced generalized superfluid model (EGSM) for level densities was selected for all the calculations. The direct channel calculations were performed by using the coupled channels model. In the case of proton induced reactions the pre-equilibrium processes were treated by the quantum mechanical multi-step direct (MSD) and the multi-step compound (MSC) models, whereas in the case of deuteron and alpha-particle induced reactions, pre-equilibrium emissions were considered by the PCROSS code. The calculations for the compound nuclear cross sections were performed by the Hauser–Feshbach theory alongwith the width fluctuation corrections based on HRTW model. In the case of EMPIRE, the optical model potentials (OMPs) of Morillon and Romain (2007) were used for proton and neutron. The OMPs for the deuteron were from Perey and Perey (1963) whereas the OMPs from McFadden and Satchler (1966) were considered for the α-particle.

TALYS is another nuclear reaction software which provides a continuous and smooth description of nuclear reactions over a wide energy and mass range. The option of back-shifted Fermi gas model was invoked to describe the continuity of the levels at higher energies. The available default models were used for the treatment of direct, pre-equilibrium and compound reactions. The default optical model potentials (OMPs) of TALYS for neutron and proton are from the local and global parameterizations by Koning and Delaroche (2003). The OMPs of Bojowald et al. (1988) were invoked for the deuteron and parameters of McFadden and Satchler (1966) were used for the α-particle. To obtain a good agreement between the nuclear theory and experiments the adjustment of nuclear model parameters was invariably necessary. The level density parameters for the concerned nuclides were slightly modified (within 10%) to get a better agreement between the calculated and measured excitation functions. In a few cases nuclear model calculations were applied to predict the results of a nuclear reaction without experimental data.

The evaluation methodology was based on the statistical treatment of the data as discussed in a few previous articles (Sudár et al., 2002; Aslam et al., 2010; Hussain et al., 2010). A polynomial function was developed for fitting the ratio of experimental cross section to the calculation by a nuclear model code (i.e. measurement/calculation). The data showing deviations larger than 3 times the standard deviation were ignored. The appropriate energy dependent normalization factor, $f(E)$, and the model calculated cross section, $\sigma_{model}(E)$, were used to derive the evaluated cross section, $\sigma_{ev}(E)$, such that

$$\sigma_{ev}(E)=f(E)\sigma_{model}(E)$$

The experimental uncertainties were also taken into account during the approximation of the polynomial function. The evaluated cross sections were generated with 95% confidence limits. This procedure was performed for each nuclear model calculation (i.e. EMPIRE and TALYS) with all the selected measurements. The best fit estimations by the nuclear model codes were then interpolated and averaged to obtain the recommended sets of cross section values with 95% confidence limits. In energy regions where no experimental data were available, extrapolations using results of nuclear model calculations were done.

3. Evaluation of production data of ^{61}Cu

The evaluated reactions are discussed below individually.

3.1. ^{61}Ni(p,n)^{61}Cu

This reaction, despite the necessity of utilizing expensive enriched ^{61}Ni as target material, is an efficient route for the