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Nuclear model analysis of excitation functions of proton and deuteron induced reactions on ^{64}Zn and ^{3}He- and α-particle induced reactions on ^{59}Co leading to the formation of copper-61: Comparison of major production routes

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HIGHLIGHTS

- Evaluation of ^{64}Zn(p,α)^{61}Cu, ^{64}Zn(d,αn)^{61}Cu, ^{59}Co(^{3}He,n)^{61}Cu and ^{59}Co(α,2n)^{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.
- Comparison of major production routes of ^{61}Cu.

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ABSTRACT

Cross section data for formation of the medically important radionuclide ^{61}Cu ($T_{½}$=3.33 h) in proton and deuteron induced reactions on enriched ^{64}Zn and in ^{3}He- and α-particle induced reactions on ^{59}Co were analyzed by using the nuclear model calculational codes, EMPIRE and TALYS. A well-defined statistical procedure was then employed to derive the recommended excitation functions, and therefrom to obtain integral yields. A comparison of major production routes of ^{61}Cu was done.

1. Introduction

The radionuclide ^{61}Cu ($T_{½}$=3.33 h) is one of the several copper radioisotopes with possible utilization in nuclear medicine. Due to its long enough half-life and appropriate decay characteristics ($I_{\beta+}$=62 %; $E_{\beta+}$=1.159 MeV; EC=38%), it is quite suitable for positron emission tomography (PET) of slower biological processes (McCarthy et al., 1999; Williams et al., 2005; Ruangma et al., 2006).

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 (Rowshanfarzad et al., 2006). Presently, (p,xn) reactions on highly enriched Ni isotopes are preferred processes for the production of ^{61}Cu (Szelecsényi et al., 1993; McCarthy et al., 1999). Recently we presented a nuclear model analysis of few possible processes for the production of ^{61}Cu from enriched Ni isotopes via proton, deuteron and α-particle induced reactions (cf. Aslam and Qaim, 2014). Alternatively, proton and deuteron induced reactions on isotopes of zinc also seem to have great potential. The use of ^{nat}Zn as target material, however, does not give high-purity ^{61}Cu for medical application due to onset of reactions leading to longer lived isotopic impurities ^{64}Cu ($T_{½}$=12.7 h) and ^{67}Cu ($T_{½}$=61.8 h). For producing high-purity ^{61}high Cu enriched ^{64}Zn needs to be employed as target material (Szelecsényi et al., 2006; Thieme et al., 2010, 2013). Furthermore, irradiations of cobalt by ^{3}He- and α-particles could also provide the desired ^{61}Cu. Those reactions have the advantage of using

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monoisotopic ^{59}Co target, though the ^{61}Cu yields are expected to be low.

In the present work we evaluated the cross section data of the following four nuclear reactions: ^{64}Zn(p,α)^{61}Cu, ^{64}Zn(d,αn)^{61}Cu, ^{59}Co(^{3}He,n)^{61}Cu and ^{59}Co(α,2n)^{61}Cu. Two nuclear model computer codes, namely EMPIRE and TALYS, were employed for consistency checks of the experimental data. The excitation function from the TENDL file (Koning et al., 2012) and the nuclear model code ALICE-IPPE (Dityuk et al., 1998) were employed, as a priori, to check the general theoretical and experimental behavior of the reactions. Different nuclear model parameters were adjusted, within their recommended limits, to obtain agreement between the theory and experiment. Nuclear model calculations were helpful in identification of reliable experimental data. The relationship established between measured and calculated cross section values was then used to generate the recommended data along with the uncertainty limits (see below). The excitation functions for the production of ^{60}Cu and ^{62}Cu were also analyzed in a few cases to assess those impurities in the production of ^{61}Cu. Those radionuclides are also potentially useful for studies of faster kinetics of molecules in PET imaging.

2. Nuclear model calculations and statistical fitting of selected data

Theoretical calculations of cross sections were carried out by using the recently developed computer codes EMPIRE 3.1 (Herman et al., 2007) and TALYS 1.4 (Koning et al., 2008). These codes incorporate various nuclear models to describe the major reaction mechanisms. The input database of both codes is based on the RIPL-3 library of the IAEA (Capote et al., 2009). The transmission coefficients were generated via the ECIS code. The optical model parameters (OMPs) for protons and neutrons used in calculations were taken from the compilation of Koning and Delaroche (2003), whereas those for α-particles were taken from McFadden and Satchler (1966). In deuteron induced reactions the OMPs of An and Cai (2006) were employed. In the TALYS code the optical model potentials for other complex particles (i.e., *t*, ^{3}He) are based on the so called folding approach. In the code EMPIRE the parameters of Becchetti and Greenlees (1969) were used for ^{3}He-particle induced reactions. The compound nuclear reactions were treated in the framework of the Hauser–Feshbach theory. The pre-equilibrium reactions were considered by two component exciton models in TALYS and by PCROSS module in EMPIRE. The level densities in the TALYS code were adopted from the back-shifted Fermi gas model, but from enhanced generalized super-fluid model (EGSM) in the EMPIRE calculations. To estimate the contribution of direct reactions the EMPIRE code employed the coupled channel model. The level density parameters were slightly adjusted to obtain a better agreement between the measured and calculated excitation functions.

A relationship was established between theory and experiment through a multistep procedure (cf. Sudár et al., 2002; Hussain et al., 2010). The cross section values were obtained from the product of a rational function and the nuclear model calculation. The evaluated cross section,$\sigma_{ev}(E)$, is described as follows:

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

where $\sigma_{mod\ el}(E)$ is the model calculated excitation function and the energy dependent normalization factor, $f(E)$, is the polynomial function which is obtained by fitting the selected measurement/calculation ratio. The data showing deviations beyond 3σ limit were neglected.

The experimental uncertainties were also taken into account during the approximation of the polynomial functions. The evaluated cross-section values with 95% confidence limits were obtained. 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 averaged and interpolated to obtain the recommended sets of cross-section values.

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

The evaluated reactions on Zn and Co targets are discussed below individually.

3.1. *^{61}Cu production from ^{64}Zn target*

The irradiation of enriched ^{64}Zn target by proton or deuteron leads to the production of ^{61}Cu. In some experiments natural zinc has also been used as target for ^{61}Cu production. However, if a proper energy control is not followed, the product ^{61}Cu may contain appreciable quantities of longer lived ^{64}Cu ($T_{½}$=12.7 h) and ^{67}Cu ($T_{½}$=61.8 h) impurities (Szelecsényi et al., 2005; 2006). The reactions on ^{64}Zn evaluated in this work are discussed below separately.

3.1.1. *^{64}Zn(p,α)^{61}Cu*

Two reactions contribute to the production of the desired radionuclide ^{61}Cu, namely (p,α) and (p,2p2n) during proton bombardment of ^{64}Zn target. The (p,2p2n) channel starts dominating above 30 MeV and no cross section measurements from enriched ^{64}Zn target are available in that energy range. Therefore this evaluation is limited only up to 30 MeV (i.e. only the (p,α) route is evaluated). In the literature two excitation function measurements with the enriched target are available (Cohen et al., 1954; Levkovskij, 1991; see Table 1). The reported data of Levkovskij (1991) were decreased by 18% in accordance with the recent suggestion by Qaim et al. (2014). The measurements on the ^{nat}Zn target (Barrandon et al., 1975; Szelecsényi et al., 2005; Uddin et al., 2007) were normalized to 100% enrichment of ^{64}Zn. Those data are shown in Fig. 1. Hermanne at al. (1999) reported cross

Table 1
Investigated nuclear reactions for the production of ^{61}Cu, Q-values and references.

Nuclear reaction	Q-value (MeV)	References
^{64}Zn(p,α)^{61}Cu	0.84	Cohen et al., 1954; Barrandon et al., 1975; Levkovskij, 1991; Hermanne et al., 1999; Szelecsényi et al., 2005; Uddin et al, 2007
^{64}Zn(d,αn)^{61}Cu	−1.38	Baron and Cohen, 1963; Williams and Irvine, 1963; Bissem et al., 1980; Bonardi et al., 2003; Tárkányi et al., 2004; Daraban et al., 2008
^{59}Co(^{3}He,n)^{61}Cu	6.61	Homma and Murakami, 1976; Michel and Galas, 1983; Jastrzebski, et al., 1986; Nagame et al., 1988; Szelecsényi et al., 2004; Fenyvesi et al., 2004; Pichard et al., 2011
^{59}Co(α,2n)^{61}Cu	−13.9	Sterns, 1962; Budzanowski et al., 1967; Zhukova et al., 1972; Homma and Murakami, 1976; Michel and Brinkmann, 1980; Gadioli et al., 1984; Jastrzebski, et al., 1986; Levkovskij, 1991; Skulski et al., 1992; Singh et al., 1993; Szelecsényi et al., 2002; Ansari et al., 2004