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Activation cross-sections of deuteron induced nuclear reactions on manganese up to 40 MeV

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ABSTRACT

In the frame of a systematic study on activation cross-sections of deuteron induced reactions experimental excitation functions on ^{55}Mn were measured with the activation method using the stacked foil irradiation technique up to 40 MeV. By using high resolution γ-ray spectrometry, cross-section data for the production of ^{56,54,52}Mn and ^{51}Cr were determined. Comparison with the earlier published data and with the results predicted by the ALICE-IPPE and EMPIRE-II theoretical codes – improved for more reliable calculations for d-induced reactions – and with data in the TENDL 2010 libraries are also included. Thick target yields were calculated from a fit to our experimental excitation curves and implications for practical applications in industrial (Thin Layer Activation) accelerator technology are discussed.

1. Introduction

Several structural components of accelerators are made of stainless steel containing around 2% of manganese. Manganese metal is also a key component of aluminum–copper alloys. For accelerator technology it is important to use low activation materials and hence reliable estimations of the activation levels are needed. Especially with the advent of new high power, high energy deuteron-accelerators installed or planned for production of high intensity neutron sources, it becomes important to look at possible activation products in these beams. The study has also importance in the frame of our systematic investigation for improvement of the presently low performance theoretical codes for deuteron induced reactions. Manganese is also a frequent alloy-element in many industrial materials that is why the possibility of application the produced radioactive isotopes for wear measurement by using Thin Layer Activation (TLA) was also in the scope of this investigation. It is especially important in the case of aluminum alloys, where iron as the best element for TLA is not present.

The element manganese has only one stable isotope ^{55}Mn. Usually there are large sets of experimental data available for activation on monoisotopic targets, but in the case of ^{55}Mn only a few authors published cross-section data for deuteron induced reactions. Gilly et al. [1], Baron et al. [2] and Coetzee et al. [3] studied the excitation functions for (d,p) reactions up to 12 MeV. Ochiai et al. [4] measured the cross-section for the activation product ^{54}Mn at 39.5 MeV in the frame of investigation of deuteron induced activation cross-section measurement for accelerator technology.

A few authors measured thick target yields for different applications: Bondarenko and Rudenko [5] used 3 MeV deuterons for activation analysis; Vakilova et al. [6] investigated the determination of elements by deuteron activation. Dmitriev et al. [7] studied the thick target yields at 22 MeV in the frame of his systematical study of a large number of target elements.

2. Experimental method and data evaluation

The experimental method used was similar to the techniques used in our numerous earlier investigations of charged particle induced nuclear reactions for different applications. Here we report only on the most salient features related to irradiation conditions, reliability of the measured data and to the used decay data and cross-section data of monitor reaction. More details can be found in the recent publications [8–10] and references to previous works herein. The excitation functions were measured by activation method using stacked foil technique. The stacks were irradiated using 21 MeV (VUB) and 40 MeV (CYRIC) incident deuteron energies. In both experiments natural high purity Ni(2)Mn(12)Cu(86)

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alloy foils (Goodfellow > 99.98%, thickness 25 μm) were assembled together with interleaved monitor Ti foils of 12 μm thickness at VUB and Al degrader and monitor foils of 100 μm at CYRIC. The Ti monitor foils were used as recoil catchers as well as for exact determination of beam intensity and energy by re-measuring the excitation function for the ^{nat}Ti(d,x)^{48}V reaction at VUB and ^{27}Al(d,x)^{22,24}Na reactions at CYRIC over the entire energy domain. The target stacks were irradiated in a Faraday-cup like target holder, equipped with a collimator (effective beam diameter on target is 5 mm) and an electrostatic secondary-electron suppressor by applying maximum +500 V on the target in order to return the secondary electrons to the target and in such a way included in the measurement. The system was calibrated before the measurement by determining the maximum voltage where the measured current does not change any more. Irradiations took place at a constant beam current of 136 nA for 30 min (VUB) and 24 nA for 30 min (CYRIC) respectively (corrected values according to monitor data).

For measurement of the produced radioisotopes in the target and the monitor foils HpGe γ-ray spectrometers were used. For energy and efficiency calibration we used ^{241}Am, ^{152}Eu, ^{137}Cs, ^{133}Ba, ^{60,57}Co, ^{54}Mn and ^{22}Na commercial standard sources. The calibration was performed before and after the measurements of the samples. Measurements of the induced activity started in Brussels shortly after EOB (End of Bombardment), while in Sendai a cooling period of at least a day was left due to the high induced activity. No chemical separation was performed and the measurements in Brussels were repeated several times up to months after EOB, allowing more accurate determination of short lived as well as long lived activation products (the short-lived information was lost in the Sendai measurements). In order to achieve good counting statistics the measuring time was chosen to reach less than 10% statistical error by the selected important peaks. To collect information also about short-lived products we made short measurements (5 min) early after the end of the irradiations with good statistics for the intense peaks from short-lived isotopes and also longer measurements later (typically 1–5 h) to improve the statistics for weak peaks from long-lived isotopes. The samples were counted at suitable large detector–sample distances (5–70 cm) to ensure low dead times (<10%) and to avoid pile-up effects. The decay and spectrometric characteristics were taken from the NUDAT2 data base [11] and are summarized in Table 1.

The *Q*-values refer to formation of the ground state and are obtained from Pritychenko et al. [12]. When complex particles are emitted instead of individual protons and neutrons the *Q*-values have to be decreased by the respective binding energies of the compound particles: np-d, +2.2 MeV; 2np-t, +8.48 MeV; n2p-^{3}He, +7.72 MeV; 2n2p-α, +28.30 MeV.

The cross-sections were calculated from the well known activation formula with measured activity, beam intensity and number of target nuclei as input parameters. Some of the radionuclides formed are the result of cumulative processes as the decay of parent nuclides contribute to the production process. The exact physical situation for the individually studied nuclides will be discussed in each case separately.

Table 1
Decay characteristics of the investigated activation products.

Nuclide	Half-life	E_γ (keV)	I_γ (%)	Contributing reaction	*Q*-value (MeV)
^{56}Mn	2.5789 h	846.75	98.9	^{55}Mn(d,p)	5.046
		1810.7	27.2		
^{54}Mn	312.05 d	834.85	99.976	^{55}Mn(d,p2n)	−12.45
^{52}Mn	5.591 d	744.23	90	^{55}Mn(d,p4n)	−33.44
		935.54	94.5	^{52}Fe decay	−36.6
		1434.1	100		
^{51}Cr	27.701 d	320.08	9.91	^{55}Mn(d,2p4n)	−39.99

As it was mentioned the beam intensity and the primary beam energy were determined on the basis of excitation function of the re-measured monitor reactions. The energy degradation as a function of depth in the stack was determined by calculation based on the Ziegler's tables [13] and corrected on the basis of the re-measured excitation function of the monitor reaction by a method described in [14].

We used the most recent upgraded recommended decay data available in the literature [15]. The excitation function of the simultaneously measured monitor reaction is shown in Fig. 1 in comparison with the recommended data. The best agreement was found after small corrections on the beam intensity (10% with respect to the Faraday cup results (see above)) and on the primary beam energy (0.5 MeV). According to the Fig. 1 the agreement is good in the whole energy range.

The uncertainty on each cross-section was estimated in the standard way [16] by taking the square root of the sum in quadrature of all individual contributions, supposing equal sensitivities for the linearly contributing different parameters appearing in the formula: counting statistics 1–18%, detector efficiency 5%, decay data 3%, effective target thickness 5% and beam current 7%. Total estimated uncertainties in the cross-sections were about 10–20%.

The contributions on the uncertainties of non-linear parameters were neglected (time, half-life, etc.). Taking into account the cumulative effects of possible uncertainties of the primary incident energy, of the thickness of the different targets and of the energy straggling the uncertainty on the median energy varies between ±0.3 MeV and ±1.2 MeV from the first to the last foil.

3. Model calculations

The updated ALICE-IPPE-D [17] and EMPIRE-D [18] codes were used to analyse and simulate the present experimental results. As described in detail in [19,20] these modified codes were developed to assure a better description of deuteron induced reactions. In the standard versions of the codes a simulation of direct (d,p) and (d,t) transitions by the general relations for a nucleon transfer probability in the continuum is included through an energy dependent enhancement factor for the corresponding transitions. The parameters were taken as described in [21]. The theoretical data from the recently corrected TENDL 2010 [22] library (based on the modified TALYS 1.2 code [23]) was used for comparison too.

4. Results

By irradiating manganese with 40 MeV deuterons, radioisotopes of Fe, Mn and Cr are produced in significant amounts. Among the radio-products formed, ^{55}Fe (2.73 y) and ^{53}Mn (3.7 x 10^8 y) have long half-lives with no gamma emission and ^{53}Fe, ^{52m}Mn and ^{55}Cr have short half-lives (<20 min) resulting in activities below the corresponding detection limits for our experimental technique under the measuring circumstances (long cooling time by high energy irradiation).

4.1. Cross-sections

The cross-sections for all the reactions studied are shown in Figs. 2–5 and the numerical values are collected in Table 2. The results of the 40 and 20 MeV primary incident energies are shown separately to see the agreement in the overlapping energy range. The reactions responsible for the production of a given activation