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Activation cross-sections of proton induced reactions on vanadium in the 37–65 MeV energy range

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ABSTRACT

Experimental excitation functions for proton induced reactions on natural vanadium in the 37–65 MeV energy range were measured with the activation method using a stacked foil irradiation technique. By using high resolution gamma spectrometry cross-section data for the production of ^{51,48}Cr, ^{48}V, ^{48,47,46,44m,44g,43}Sc and ^{43,42}K were determined. Comparisons with the earlier published data are presented and results predicted by different theoretical codes (EMPIRE and TALYS) are included. Thick target yields were calculated from a fit to our experimental excitation curves and compared with the earlier experimental yield data. Depth distribution curves to be used for thin layer activation (TLA) are also presented.

1. Introduction

Many authors published experimental cross-section data for proton induced reactions on natural vanadium, especially at lower energies in the frame of basic nuclear physics research [1–4]. A few works reported experimental thick target yields [5–8]. As mentioned, most of the literature cross-sections data were obtained in the low energy region (below 30 MeV incident energy) and only a few (in some cases contradictory) data sets were measured at higher energy. We participated in an IAEA-CRP setting-up an activation data library for proton and deuteron induced reactions for the FENDL project. Vanadium was included into this library as an important construction and alloying material in accelerator technology. During compilation of the experimental data and comparison with the TENDL library it was recognized that the activation data at higher energies are contradictory and the theoretical model description for radionuclides far from the target mass number is very poor. The maximum proton energy presently available for us was 65 MeV at the Cyclone110 cyclotron of Louvain la Neuve (LLN) Belgium through the HAS-FWO (Vlaanderen) collaboration. We decided therefore to re-measure the cross-section data for all possible γ-emitting activation products of ^{nat}V in the 37–65 MeV proton energy range.

Natural vanadium consists of two isotopes: for more than 99.75% it is formed by stable ^{51}V while the very long-lived ^{50}V ($T_{1/2}$ = 1.4 10^{17} a) occurs only in 0.25%. Experimental data on ^{nat}V can hence be interpreted in broad energy range as coming from a monoisotopic ^{51}V target and reaction cross-sections can be derived.

Among the reaction products ^{51}Cr is an attractive radionuclide for use in diagnostic nuclear medicine due to its favorable nuclear characteristics such as a half-life of 27.7010 days and decay by electron capture, resulting in emission of gamma photons with energy of 320.0824 keV (9.91%). Its main applications is labeling of red blood cells for measurement of mass or blood volume, labeling of platelets to determine their survival time as well as in sequestration studies for the diagnosis of gastrointestinal bleeding [9]. The favorite routes for no carrier added production are the (p, n) and (d,2n) reactions on vanadium [10,11].

Among the scandium radionuclides ^{44}Sc, ^{47}Sc and ^{46}Sc have been used in biological and medical studies. The ^{47}Sc has attracted attention because of its favorable decay characteristics (half-life: 3.35 d; average electron energy: 162 keV; E_γ: 159 keV) for therapeutic application and for SPECT imaging. The applicability of ^{44g}Sc as a matching pair for ^{47}Sc through PET imaging, ^{44m}Sc for radionuclide therapy. The longer-lived ^{46}Sc was used for regional blood flow studies and investigation of radiochemistry of scandium. Recently ^{46}Sc has been advocated as an additional monitor for proton and deuteron reactions and will be taken up in the recommended data base of the IAEA-CRP [12]. The main

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charged particle production routes to gain carrier free Sc radioproducts include proton and deuteron induced reactions on calcium, titanium and vanadium.

^{43}Sc was shown to be an excellent tool for studying blood perfusion in the heart and for diagnosing myocardial infarcts. The main production routes at charged particle accelerators are the ^{40}Ar(α,p), ^{nat}Ca(p,x) or ^{nat}Ca(d,x) and high energy reactions on Ti and V, but in these last cases the cross sections are small.

2. Experimental and data processing

The excitation functions for the ^{nat}V(p,x) reactions were measured via an activation method by using the stacked foil irradiation technique followed by high resolution γ-ray spectrometry without chemical separation.

The irradiation was performed at the LLN Cyclone110 cyclotron with 65 MeV protons for 1 h at 35 nA beam intensity. The stack contained a 19 times repeated set of 10 μm Al, 116 μm In, 99.2 μm Al, 8.41 μm V, 99.2 μm Al, 26.2 μm Ho and 99.2 μm Al foils. The Al-monitors were also used as recoil catchers as well as for exact determination of beam intensity and energy by re-measuring the excitation function for the ^{27}Al(p,x)^{22,24}Na reactions over the entire energy domain [13]. The target stacks were irradiated in a Faraday-cup like target holder, equipped with a long collimator (effective beam diameter on target is 5 mm).

The gamma activity of the produced radionuclides was measured with standard high purity Ge detectors at the Cyclotron Laboratory of VUB-Brussels.

Measurements of the induced activity started about 20 h after EOB (End of Bombardment), hence only activation products with a half-life longer than 3 h could be assessed. The samples were counted repeatedly at varying sample detector distance for weeks after EOB. The evaluation of gamma-spectra was made by automatic peak recognition programs and in a manually controlled interactive way.

For most of the radionuclides assessed different independent γ-lines are available, allowing an internal check of the consistency of the calculated activities.

The cross-sections were calculated by using the well-known activation formula with input parameters: measured activity, particle flux and number of target nuclei. In this study only a couple radionuclides formed as a result of a cumulative process by decay of a parent nuclide. The exact physical situation will be discussed individually for each activation product.

The decay and spectrometric characteristics, needed to transform count rates to activity of the different activation products at EOB, were taken from the NUDAT2 data base [14] and are summarized in Table 1. This table also includes the reaction Q-values of the contributing reactions [15].

The number of incident particles was initially derived from continuously measured beam current on target. The mean energy in each target foil was estimated by a stopping calculation from the incident beam energy and target thickness [16].

The beam energy and intensity parameters were further adapted by taking into account the comparison of the excitation function of the ^{27}Al(p,x)^{22,24}Na reactions, re-measured over the whole energy domain studied, with the recommended values in the updated version of IAEA-TECDOC 1211 [17].

The uncertainties of the median energies were estimated from the uncertainties of the cumulative contributing processes (taking into account possible incident energy variation, thickness variation of the different targets and straggling effects) resulting in around ±0.3 MeV for the first foil and about ±1.3 MeV for the last foil.

The uncertainty on each cross-section point was estimated in the standard way [18] taking the square root of the sum in quadrature of all individual contributions, supposing equal sensitivities for the different parameters appearing in the formula. The following individual uncertainties are included in the propagated error calculation: absolute abundance of the used γ-rays (5%), determination of the peak areas including statistical errors (4–10%), the number of target nuclei including non-uniformity (5%) and detector efficiency (10%). The total uncertainty of the cross-section values was estimated to be 8–14%. The strongly non-linear effect of the possible uncertainty of the half-lives, cooling time and measuring time for samples was not taken into account.

3. Model calculations

The cross-sections of the investigated reactions were compared with the data given in the last two on-line TENDL libraries to show the development of the predictions (from TENDL-2010 to TENDL-2014 and TENDL-2015) [19]. These libraries are based on both default and adjusted TALYS calculations Koning and Rochman [20]. For comparison also the results of the latest version of the EMPIRE code (EMPIRE 3.2 (Malta)) are presented [21,22]. Default input was used in the most cases, expect the TENDL libraries, where the TALYS results are adjusted in an unknown (for the public user) way.

4. Results

The cross-sections for all the reactions studied are shown in Figs. 1–11 and the numerical values are collected in Table 2. No practical contribution from the reaction on the low abundance ^{50}V can be distinguished, hence all results are practically equivalent to reaction cross sections on ^{51}V.

4.1. Production of ^{51}Cr

Many experimental data sets exist for production of ^{51}Cr ($T_{1/2}$ = 27.701 d) especially at low energies for basic research. Results were published by Tanaka, Albert, Shore, Taketani, Wing, Hansen, Albouy, Hontzeas, Humes, Johnson 1964, Dell, Harris, Chodil, Johnson 1958, Gadioli, Barandon, Mehta, Michel 1979, Michel 1980, Zyskind, Kailas, Bastos, Levkovskij (normalized), Jung, Musthafa and Carlson [1,2,23–46]. The data differ by a factor of two around the maximum. A recommended data set was also proposed earlier in the IAEA TLA library [47]. In the presently investigated energy range experimental data are available from five groups (Albuoy, Michel 1983, Michel 1979, Hontzeas and Zhao) [29,30,37,48,49]. Our data are in good agreement with the earlier results of Michel and Zhao (Fig. 1). Both the TENDL and the EMPIRE predictions describe the experimental data acceptable well.

4.2. Production of ^{48}Cr

For production cross sections of ^{48}Cr ($T_{1/2}$ = 21.56 h) the earlier experimental data of (Heininger, Hontzeas and Michel 1979) [30,37,50] show large discrepancies (Fig. 2). Our data support the results of the Hannover group Michel et al. [48]. The TENDL prediction shows only a moderate agreement (lower maximum, energy shift). A gradual change (increase) of the subsequent TENDL versions can be observed. The EMPIRE 3.2 results describe both the maximum and the magnitude of the excitation function well.

4.3. Production of ^{48}V

The measured activation cross sections for ^{48}V ($T_{1/2}$ = 15.9735 d) formation are cumulative. They include the direct production by (p,p3n) reaction with a threshold around 32 MeV and indirect production through the decay of the ^{48}Cr (21.56 h) parent isotope. The agreement with the earlier experimental data of Heininger,