DEUTERON ACTIVATION OF CHROMIUM FOR THIN LAYER ACTIVATION STUDIES

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The present study is aimed at the technological interest of using the thin layer activation technique (TLA) for surface performance evaluation of chromium based materials. ^{51}Cr, from the deuteron induced reaction in natural chromium, has been selected as a tracing element and its relative activity as a function of depth has been considered. Results are based on calculations using computer codes, STAPRE and ALICE and experimental verifications, using the stacked foil technique and electropolishing of activated bulk samples. A very good agreement of the various methods was obtained. It is shown that using 10 MeV deuterons, a reasonable homogeneous activation over 120 μm can be obtained.

1. Introduction

The wide use of chromium in the form of protective coatings and as alloying element is mainly due to its excellent resistance against corrosion. Recent technological developments have also made possible the application of components with chromium as the main material constituent [1]. Due to increasingly greater demands imposed on materials, advanced testing techniques are employed to establish their applicability. A very sensitive technique to measure material degradation from such processes as wear and corrosion, is that of Thin Layer Activation (TLA). The method comprises of generating at least one radionuclide in the material component by activation with charged particles and monitoring the loss of activity of the various radionuclides due to material loss [2,3].

To relate the technically relevant parameters in surface degradation, e.g. loss in material thickness, to the γ-activity, which is normally monitored, the distribution of the induced γ-activity versus depth should be known. The largest use of TLA is in the field of wear testing of iron based materials, such as steels. Mostly activation of iron with protons is used and the relevant reactions are rather well known and were recently compiled [4]. There exists a lack of data for the activation of various other elements. For the case of chromium, the production of the radionuclide ^{51}Cr by deuteron activation of natural chromium has been recently studied [5]. For TLA purposes this radionuclide is of interest, because of its half-life of 27.7 days and specific γ-radiation of 320 keV. Additionally, the fact that this isotope is of the same chemical element as the target, makes it suitable for those applications where element selective surface degradation occurs, as e.g. in various corrosion processes.

^{51}Cr can be produced by a (p, pn) reaction on ^{52}Cr, however in this case a large amount of ^{52}Mn ($T_{1/2}$= 5.6 days) is formed via (p, xn) reactions on ^{52}Cr and ^{53}Cr. This leads to interfering high background radiation in the γ-spectrum of an activated natural chromium sample. Production of ^{51}Cr by deuteron activation of natural chromium avoids the formation of ^{52}Mn, this is possible by selecting the incident deuteron energies below the threshold energy of (d, xn) reactions. In Table 1 the deuteron induced reactions leading to ^{51}Cr, in the energy range of 0 to 18 MeV, are reported.

Table 1: Deuteron induced reactions for ^{51}Cr production (between 0 and 18 MeV), isotopic abundance θ, threshold energies E_{Th} and Coulomb barrier energies E_{Coul}.

Induced Reaction	θ	E_{Th} (MeV)	E_{Coul}(MeV)
^{50}Cr(d,p)^{51}Cr	4.35	0	5.15
^{52}Cr(d,p2n)^{51}Cr	83.79	14.8	7.8
^{50}Cr(d,n)^{51}Mn		0	5.15
^{51}Mn $\beta^+ \rightarrow$ ^{51}Cr			

In the present study the cross section data obtained by computer calculation and stacked foil technique, as well as electropolishing of activated bulk samples, have been used to obtain the calibration curve, γ-activity versus depth, which is suitable for TLA application.

2. Experimental

The activation yield for the deuteron induced production of ^{51}Cr as a function of the energy, in the range of 0 to 18 MeV, was measured experimentally via the stacked foil technique, using a MC-40 variable energy cyclotron. High purity (≥99.99%) natural chromium foils of 10±1 μm thickness and 10mm diameter on 125μm mylar support were used in these experiments. The targets were prepared by stacking a maximum of 10 chromium foils, alternated

with aluminium foils of different thickness, starting from 100 μm to 13 μm, in order to have a beam energy reduction of about 1.5 MeV on each chromium foil. The energy degradation of the beam, passing through the various absorbers, was calculated from the energy projected range data output of the TRIM code [6]. The ^{51}Cr activity induced in each chromium foil was measured by γ-spectrometry using a germanium detector connected with a computerised analysing system. In Fig. 1, the γ-spectrum of an activated chromium foil shows a specific γ-radiation at 320 keV, ^{48}V and ^{54}Mn respectively arise from the (d, α) reaction on ^{50}Cr and from the (d, n) reaction on ^{53}Cr.

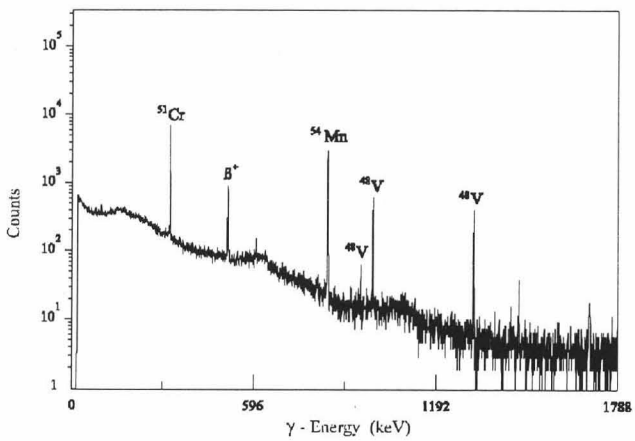

Fig. 1: γ-Spectrum of a deuteron irradiated (7 MeV) natural chromium sample.

Additional experimental data were obtained by activation of bulk samples of natural chromium (99.7%) with 10 MeV deuterons. Layers of the material were successively electrochemically removed. This was carried out in a 6-2-1-1 solution of sulphuric acid, phosphorous acid, citric acid and water at 25 °C. A lead sheet served as the counter electrode. A potential difference of 15 V and a current density of 2500 A/m^2 was employed. These conditions led to an even removal of the material with a dissolution rate of approximately 1 μm/min, which was determined by thickness measurements using a micrometer with an accuracy of 1 μm. After the dissolution of each layer, the ^{50}Cr-activity remaining in the bulk sample was measured using the same experimental set up and conditions as used for the foils.

3. Results and discussion

The experimental results of the stacked foils were compared with calculations performed using two computer codes, STAPRE [7] and ALICE [8], based on the pre-equilibrium statistical compound model, for the calculation of nuclear cross sections (σ(E)). Both codes were employed for the computation of the total cross section for the production of ^{51}Cr via deuteron activation. Once the activation cross sections are known, the theoretical thin target yield (Y(E)) can be calculated by using expression (1):

$$Y(E) = K \cdot \int_{E_{threshold}}^{E_{incident}} \frac{\sigma(E)}{S(E)} dE \qquad (1)$$

where K is a constant depending on the chemical and physical nature of the target material and S(E) is the stopping power of the incident particle in the material, which was calculated using the code TRIM.

Fig. 2 shows the theoretical and experimental yield as a function of energy.

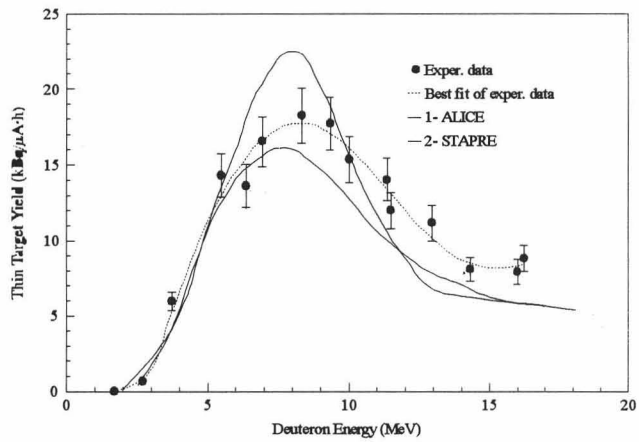

Fig. 2: Activation yield for deuteron induced production of ^{51}Cr

The uncertainties in the activation yield of the experimental data are related to uncertainties in foil thickness, counting statistics, detection efficiency, calibration and current integration, resulting in a standard deviation of 10.6%. The uncertainty in the energy value is mainly due to the uncertainty in the stopping power data by TRIM and to those of ΔE/E of the incident beam and of the absolute value of the beam energy, resulting in a standard deviation of 3.3%. The observed differences between the maximum yields calculated by STAPRE and ALICE as well as the energy shift of about 1 MeV arise from the parameters used for the codes, since for ALICE the default parameters have been used. Comparing these curves with the experimental values, the best agreement is found with the STAPRE output. Especially at lower energies the agreement is satisfying. A maximum yield value is reached at around 8 MeV.

From these results the optimum energy for TLA applications was estimated to be 10 MeV, corresponding to a yield of approximately 90% of the maximum value. This is expected to give a reasonable depth of activation as well as a rather homogeneous activation over this depth. The latter case makes quantification of material loss relatively easy, since it is directly related to activity loss.

The cross section data obtained by computer calculations and using the stacked foil technique have been used to obtain the calibration curve which is suitable for TLA studies. The range of deuterons in chromium as a function of the particle energy has been calculated using the TRIM code. These data and the data obtained on the

activated bulk chromium samples are presented in Fig. 3 as relative activity (i.e. the activity of the whole sample is defined as 100%) versus depth.

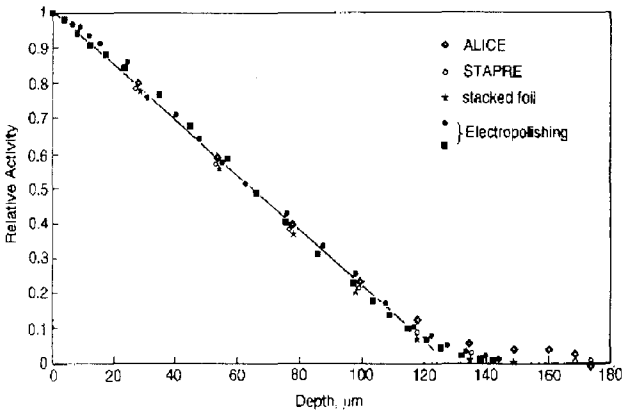

Fig.3: Total ^{51}Cr activity versus depth for 10 MeV deuterons in natural chromium

4. Concluding remarks

A very good agreement of the various methods, for the determination of the relative activity versus depth, has been obtained (Fig. 3), despite a rather large difference in the absolute yield data (Fig. 2). This result suggests the possibility of employing computer codes to assess the optimal experimental activation conditions: type of particle, incident energy, beam intensity and irradiation time required for material studies employing TLA. The activation of natural chromium, using 10 MeV deuterons, provides a quite homogeneous activity distribution over 120 µm which is a very useful depth for the application of the TLA for an engineering point of view.

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