Production of $^{55}$Co via the $^{54}$Fe($d, n$)-process and excitation functions of $^{54}$Fe($d, t$)$^{53}$Fe and $^{54}$Fe($d, \alpha$)$^{52m}$Mn reactions from threshold up to 13.8 MeV

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Summary. For production of the medically interesting $\beta^+$-emitter $^{55}$Co ($T_{1/2} = 17.6$ h) via the $^{54}$Fe($d, n$)-reaction, 91.6% enriched $^{54}$Fe$_2$O$_3$, mixed with Al powder, was pressed to a pellet which could be irradiated with 14 MeV deuterons at 4 $\mu$A in a water-cooled target system. A separation method was developed which led to $>99.9\%$ pure $^{55}$Co and allowed a recovery of the enriched target material. For a target thickness of $E_d = 12.6 \rightarrow 5$ MeV, the experimental thick target yield of $^{55}$Co after chemical separation amounted to about 13 MBq/$\mu$A·h, which is about 60% of the theoretical value. In a 3 h irradiation at 4 $\mu$A, the batch yield of $^{55}$Co achieved was 160 MBq (4.3 mCi). An 8 h irradiation could lead to a batch yield of about 400 MBq. The $^{54}$Fe($d, n$) reaction leads to the highest purity product, provided highly enriched $^{54}$Fe is used as target material [8, 9]. A medium-scale production of $^{55}$Co via this route has been described [8]. We investigated this reaction earlier [9] from the viewpoint of nuclear data. Now we describe some work on targetry and chemical processing. Cross sections for the competing ($d, t$) and ($d, \alpha$) reactions on $^{54}$Fe are also reported.

2. Production of $^{55}$Co

2.1 Targetry

About 125 mg of $^{54}$Fe$_2$O$_3$ (stated isotopic composition: $^{54}$Fe = 91.6%, $^{56}$Fe = 8.20%, $^{57}$Fe = 0.2%; stated chemical impurities: Mg, Al, Cl, Co, Ni, Cu, Sb (< 50 ppm); Si, Ca, Cr (< 300 ppm), supplied by Chemotrade GmbH, Germany/Russia) was thoroughly mixed with 45 mg of fine Al powder (> 98%, < 160 $\mu$m, FLUKA, Germany) in a mortar, and then pressed to a pellet of 1.3 cm diameter at 10 tons/cm$^2$ (23.8 MPa). Addition of Al was necessary to be able to press $^{54}$Fe$_2$O$_3$ to a pellet. The thickness of the pellet amounted to about 250 $\mu$m. The pellet was placed in the groove of a Cu-target holder and covered by a 100 $\mu$m Cu foil. The target holder was then attached to an irradiation system which was cooled from three sides by flowing water at 13 °C. This target arrangement [cf. 10] could withstand an irradiation with 14 MeV deuterons at 4 $\mu$A. The effective deuteron energy in the target pellet was 12.6 $\rightarrow$ 5 MeV.

2.2 Chemical processing

After irradiation the pellet was dissolved in 4 mL of conc. HCl and 2 mL of H$_2$O$_2$. The solution was evaporated to dryness to remove H$_2$O$_2$, the residue was taken up in 5 mL of conc. HCl and $^{54}$Fe was removed by extraction with two 10 mL portions of diethyl ether saturated with conc. HCl. The aqueous solution was evaporated to dryness slowly to remove traces of ether. The residue was taken up in 10 mL of conc. HCl. It contained radiocobalt and radiomanganese at the no-carrier-added level and Al in macroquantities. It was enriched $^{58}$Ni as target material, about 0.5% $^{57}$Co impurity ($T_{1/2} = 271.3$ d) cannot be avoided [7]. The $^{54}$Fe($d, n$)$^{55}$Co reaction leads to the highest purity product, provided highly enriched $^{54}$Fe is used as target material [8, 9]. A medium-scale production of $^{55}$Co via this route has been described [8]. We investigated this reaction earlier [9] from the viewpoint of nuclear data. Now we describe some work on targetry and chemical processing. Cross sections for the competing ($d, t$) and ($d, \alpha$) reactions on $^{54}$Fe are also reported.

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then transferred to a column (Ø = 2.0 cm, height = 10 cm) filled with the anion-exchange resin Dowex 1 × 8, chloride form, 100–200 mesh (FLUKA). The column was conditioned with 10 mL of conc. HCl, then eluted with 10 mL of 6 M HCl whereby radiomanganese was quantitatively removed. Thereafter elution was done with 60 mL of 4M HCl; radiocobalt was removed from the column and existed in about 40 mL of the elute. AI remained on the column. The fractions containing 55Mn and 55Co were separately collected and concentrated by evaporation. 55Co was finally obtained as 55CoCl2 in 2 mL of 1 M HCl. The recovery of radionuclidic purity of the separated radiocobalt was determined via high-resolution HPGe detector γ-ray spectrometry. The composition was found to be: 55Co (> 99.9%), 56Co (< 0.02%), 57Co (0.06%). The level of 55Mn was < 0.01%. These results are in general agreement with the production data reported by Sharma et al. [8] using 87.9% enriched 54Fe targets. The 56,57Co impurities are due to the presence of 56Fe and 57Fe in the enriched 54Fe target material. The radionuclidic purity of 55Co produced via the 54Fe(d, n) process using 91.6% enriched 54Fe is not as good as from the 99.85% enriched 54Fe used earlier [9], but it is still very good.

The chemical purity of the separated radiocobalt was checked by ICP-OES. The amounts of Fe, Mn, Ni, Cr and Mg were found to be < 1 ppm. The amount of Al was estimated via neutron activation analysis using the nuclear reaction 27Al(n, p)27Mg (T1/2 = 9.5 min) with 14 MeV d(He) neutrons. An upper limit for the Al content in the separated radiocobalt was placed at 50 ppm.

### 2.4 Prospects of availability and use of 55Co

The 56Fe(p, 2n)55Co reaction gives the highest 55Co yield and, in spite of the rather high 56Co impurity, the product has been used in a few limited brain studies on patients [cf. 4]. The use of 56Fe as target material makes targetry easy and 55Co quantities of about 2 GBq can be conveniently produced. However, the 56Co impurity limits the use of the product only over short periods, i.e. for fast metabolic processes. The 55Ni(p, α)55Co process is technically well developed [cf. 7] but the yield of 55Co is low and the level of 56Co impurity (0.5%) is of some concern. It can thus probably be used only in animal experiments. The radionuclidic purity of 55Co produced via the 54Fe(d, n) reaction using 91.6% enriched 54Fe is still superior to the products from the other two routes. Based on the cross sections for the 54Fe(d, n)55Co and 58Fe(d, 2n)56Co reactions [cf. 11] we estimated the impurity levels of 57Co and 56Co for various enrichments of 54Fe. For an 54Fe enrichment of 86% and the energy range Е = 10 → 5 MeV, the levels of 57Co and 56Co in 55Co would amount to ∼ 0.11 and 0.04%, respectively. In order to keep the level of long lived impurities below 0.1%, it is thus essential that 54Fe enrichments lower than 90% are not used. The availability of high-purity 55Co might lead to its use over more extended periods. The major difficulty with the 54Fe(d, n) process, however, relates to targetry. On the one hand, the cost of enriched 54Fe is rather high and, on the other, no high-current irradiation technique has been reported. To date the yields are limited to about 400 MBq. For a wider use of this production route, development of high-current targetry is absolutely necessary.

### 3. Cross section measurements

Detailed cross section measurements on the 54Fe(d, n)55Co and 54Fe(d, α)55Mn reactions have already been described [cf. 9 and references cited therein]. However, in contrast to the short-lived products formed via the 54Fe(d, α)52mMn and 54Fe(d, t)53m,56Fe processes were not investigated. There is some interest in investigating those products. The radionuclide 52mMn (T1/2 = 21.1 min; Еα = 2.6 MeV; Iγ = 97%; Еγ = 1434.1 keV; Iγ = 98.2%) is potentially useful for PET studies on fast manganese uptake in an organ. The radionuclides 53mFe (T1/2 = 2.6 min; IT = 100%; Еγ = 1328.2 keV; Iγ = 86.0%) and 58Fe (T1/2 = 8.5 min; Еγ = 2.8 MeV; Iγ = 97%; Еγ = 377.9 keV; Iγ = 42%) have relatively high nuclear spins (19/2− and 7/2− respectively). Their formation at a relatively low excitation energy is thus of fundamental interest.

As usual, cross sections were measured by the well-known stacked-foil technique. About 25 μm thick 56Fe foils were irradiated in the form of several stacks. The irradiation time in each case was 15 min and the radioactivity was determined non-destructively via high-resolution HPGe detector γ-ray spectrometry. The 8.5 min 58Fe was identi-
fied via the 377.9 keV γ-ray and the 21.1 min $^{52m}$Mn via the 1434.1 keV γ-ray. Other techniques related to deuteron flux measurement, determination of absolute activity and calculation of cross section and its error were similar to those described earlier [9]. For deuteron beam monitoring the reaction $^{54}$Fe$(d, xn)^{53}$Co was used. Its cross sections were taken from a recent evaluation [12].

The results are given in Table 2 and have been corrected for the 5.8% abundance of $^{54}$Fe in nat Fe. At the deuteron energies involved in this work, reactions on the most abundant isotope $^{56}$Fe cannot contribute to the formation of $^{52m}$Mn and $^{53}$Fe. Despite an intensive and careful γ-ray spectrometric analysis the very high-spin and short-lived $^{53}$Fe ($T_{1/2} = 2.6$ min) could not be identified. An upper limit for its formation cross section at $E_d = 13.8$ MeV was placed at 0.1 mb. Other results are discussed below.

### 3.1 $^{54}$Fe$(d, t)^{53}$Fe reaction

The excitation function is shown in Fig. 1. The experimental threshold of the reaction is at about 10 MeV. As expected, the cross section increases with the increasing deuteron energy. The product could be formed, in principle, via three routes, viz. $^{54}$Fe$(d, p2n)^{53}$Fe, $^{54}$Fe$(d, dn)^{53}$Fe and $^{54}$Fe$(d, t)^{53}$Fe processes. The Q-values of those processes are $-15.61$, $-13.38$ and $-7.12$ MeV, respectively. It is therefore obvious that over the energy region considered here, the $(d, t)$ process makes the largest contribution. Since the deuteron could easily pick up a neutron, the emission of the triton would presumably occur via a direct interaction process.

The formation of $^{53}$Fe has been investigated in $^{54}$Fe$(p, pn)^{53}$Fe [13], $^{54}$Fe$(n, 2n)^{53}$Fe [14], $^{52}$Cr$(^3$He,2n)$^{53}$Fe [15] and $^{54}$Fe$(d, t)^{53}$Fe [this work] processes. Their cross sections at an excitation energy of about 4 MeV above the respective threshold amount to 60, 70, 32 and 0.5 mb, respectively. The lowest value for the $(d, t)$ reaction depicts that this process is intrinsically weak.

### 3.2 $^{54}$Fe$(d, α)^{52m}$Mn reaction

The excitation function of this reaction is shown in Fig. 2. The threshold lies at about 4.0 MeV and the maximum at about 10.5 MeV. The shape of the curve is similar to those of other $(d, α)$ reactions in this mass region. Clark et al. [16] had described a curve for this reaction; however, without giving any data points or errors. Their curve is reproduced in Fig. 2. We believe our data to be more accurate because of the use of improved experimental techniques, especially with regard to charged particle flux measurement and high-resolution γ-ray spectrometry.

### 3.3 Cross section ratios for the isomeric pair $^{52m,α}$Mn

Based on the cross sections of the $^{54}$Fe$(d, α)^{52m}$Mn reaction measured earlier [9] and the data for the $^{54}$Fe$(d, α)^{52m}$Mn reaction determined in this work, the ratio $\sigma_{m}/\sigma_{g}$ was cal-

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**Table 2.** Measured cross sections of some deuteron induced nuclear reactions.

<table>
<thead>
<tr>
<th>Deuteron energy (MeV)</th>
<th>$^{54}$Fe$(d, α)^{52m}$Mn</th>
<th>$^{54}$Fe$(d, t)^{53}$Fe</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.6 ± 0.5</td>
<td>8 ± 1.5</td>
<td></td>
</tr>
<tr>
<td>6.0 ± 0.5</td>
<td>18 ± 3</td>
<td></td>
</tr>
<tr>
<td>6.5 ± 0.5</td>
<td>20 ± 3</td>
<td></td>
</tr>
<tr>
<td>7.2 ± 0.5</td>
<td>23 ± 3</td>
<td></td>
</tr>
<tr>
<td>7.6 ± 0.4</td>
<td>26 ± 3</td>
<td></td>
</tr>
<tr>
<td>8.8 ± 0.4</td>
<td>29 ± 3</td>
<td></td>
</tr>
<tr>
<td>9.8 ± 0.4</td>
<td>33 ± 4</td>
<td></td>
</tr>
<tr>
<td>10.3 ± 0.4</td>
<td>33 ± 4</td>
<td></td>
</tr>
<tr>
<td>10.7 ± 0.4</td>
<td>34 ± 4</td>
<td></td>
</tr>
<tr>
<td>11.2 ± 0.3</td>
<td>32 ± 4</td>
<td>0.17 ± 0.06</td>
</tr>
<tr>
<td>11.6 ± 0.3</td>
<td>30 ± 4</td>
<td>0.28 ± 0.09</td>
</tr>
<tr>
<td>12.1 ± 0.3</td>
<td>26 ± 3</td>
<td></td>
</tr>
<tr>
<td>12.5 ± 0.3</td>
<td>26 ± 3</td>
<td></td>
</tr>
<tr>
<td>13.1 ± 0.3</td>
<td>24 ± 3</td>
<td>0.36 ± 0.09</td>
</tr>
<tr>
<td>13.6 ± 0.3</td>
<td>22 ± 3</td>
<td></td>
</tr>
<tr>
<td>13.8 ± 0.3</td>
<td>24 ± 3</td>
<td>0.49 ± 0.14</td>
</tr>
</tbody>
</table>

a: The deviation gives a quadratic sum of the primary energy error and the energy spread within a sample.

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**Fig. 1:** Excitation function of the $^{54}$Fe$(d, t)^{53}$Fe reaction. The solid line is an eye-guide.

**Fig. 2:** Excitation function of the $^{54}$Fe$(d, α)^{52m}$Mn reaction. The solid line is an eye-guide. The dashed line gives the results of Clark et al. [16].
calculated and the results are shown in Fig. 3. Because of appreciable difference in the half-lives of the two isomers, measurements were done in two experiments, one involving short irradiations and the other longer irradiations. The ratio is high at low incident deuteron energies but decreases rapidly with the increasing energy. The trend is explainable in terms of the spins of the two states involved. The metastable state with a lower spin (2<sup>+</sup>) is formed favourably at low incident projectile energies. The higher spin ground state (6<sup>+</sup>) is more favoured at high projectile energies.

The σ<sub>α</sub>/σ<sub>d</sub> for the isomeric pair 52m,g Mn has also been determined in the 40<sup>Cr</sup>(p, xn)52m,g Mn and 54<sup>Fe</sup>(d, α)52m,g Mn as a function of the respective projectile energy. The results for the former reaction have been taken from Klein et al. [17] and those for the latter reaction are based on an earlier [9] and present measurements. The solid lines are eye-guides.

**References**