PRODUCTION OF ^{155}Tb FOR NUCLEAR MEDICINE IN THE REACTIONS ^{155}Gd(pn), ^{156}Gd(p2n), AND ^{155}Gd(d2n)

P. P. Dmitriev, G. A. Molin,
Z. P. Dmitrieva

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In producing a radionuclide for medicine, one must ensure the required purity and the possibility of producing the nuclide in quantities sufficient for practical use.

In [1] the yields [in units MBq/(μA·h)] are given for the nuclides ^{155}Tb (with a half-life of 5.32 days), ^{151}Tb (with a half-life of 17.6 h; decays to ^{151}Gd, with a half-life of 120 days), ^{152}Tb (17.5 h), ^{153}Tb (2.34 days; decays to ^{153}Gd, with a half-life of 242 days), ^{154}Tb (21.4 h), and ^{156}Tb (5.34 days) upon bombardment of a thick europium target with 42-MeV alpha particles: ^{153}Eu(α2n)^{155}Tb 4.8 ± 1.1; ^{151}Eu(α4n)^{151}Tb 1.8; ^{151}Eu(α3n)^{152}Tb – 21.8; ^{151,153}Eu(α; 2n, 4n)^{153}Tb – 10.5 ± 1.7; ^{151,153}Eu(α; n, 3n)^{154}Tb – 22; and ^{153}Eu(αn)^{156}Tb – 0.48 ± 0.7.

When a thin target of ^{153}Eu enriched to approximately 98% is bombarded with ~34-Mev alpha particles, the ^{155}Tb yield increases by 40 to 60% (depending on target thickness). At the end of bombardment, the impurities are as follows: ^{152}Tb∿19%, ^{153}Tb∿1.6%, ^{154}Tb∿32%, ^{156}Tb∿1.9%. After holding for about 8 to 10 days, in effect only the impurities ^{153}Tb∿0.12% and ^{156}Tb∿2% will remain, but the actual yield of ^{155}Tb will decrease by a factor of approximately 3.7, to approximately 1.3 MBq/(μA·h).

The object of this work is to show that when enriched isotopes of gadolinium are bombarded with protons and deuterons, the ^{155}Tb yield will be much higher and the total impurity content of other isotopes of terbium will be lower than those given for ^{153}Eu.

The ^{155}Tb and ^{156}Tb yields were measured during bombardment of thick gadolinium targets with protons and deuterons having an energy of approximately 11 to 22 MeV. We bombarded Gd_2O_3 samples in the deflected beam of the cyclotron at the FÉI [I. V. Kurchatov Physics and Energy Institute] (the conversion factor to pure gadolinium was 1.29). The radionuclides were identified by the gamma-ray energy and the half-life. The activity of ^{155}Tb and ^{156}Tb were determined from the photopeaks of selected gamma lines: ^{155}Tb 105.3 keV, 18%, and ^{156}Tb 1222 keV, 29.4%. For the gamma-ray line at 105.3 keV we introduced a correction for self-absorption in the Gd_2O_3 sample. The gamma-ray energy and the quantum efficiency and the half-lives of the nuclides were taken from [2]. The photopeaks were measured with a gamma spectrometer using a DGDK-50Al Ge(Li) detector approximately 20 days after bombardment. Copper monitoring foils were used to measure the integrated bombardment current. The method used to measure the activity of the isotopes and the integrated bombardment current is analogous to the one described in [3]. The error of measured yields was 14-17% for ^{155}Tb and 12-15% for ^{156}Tb, and was basically due to systematic errors in measurements of the activity of ^{155}Tb and ^{156}Tb and the integrated bombardment current.

The results of measurements of the ^{155}Tb and ^{156}Tb yields are shown in Fig. 1 for six values of the particle energy. We can see that for a particle energy of 22 MeV, for protons the yield [in MBa/(μA·h)] is 15.2 for ^{155}Tb and 13.3 for ^{156}Tb; for deuterons, the figures are 13.7 for ^{155}Tb and 14.2 for ^{156}Tb. Here, other long-lived terbium nuclides also are formed. Theoretical calculations and estimates for the yields of analogous nuclear reactions [4] give the following yields [MBq/(μA·h)]: for proton bombardment of gadolinium, 0.79 for ^{151}Tb and 0.84 for ^{152}Tb; 4.2 for ^{153}Tb and 16 for ^{154m}Tb (24.0 h); 36 for ^{154g}Tb; 60 for ^{156m}Tb (24.4 h); 0.0025 for ^{157}Tb (150 years); 0.00044 for ^{158}Tb (150 years); and 0.045 for ^{160}Tb (72.3 days). For deuteron bombardment of gadolinium, the figures are 0.23 for ^{151}Tb, 0.66 for ^{152}Tb; 0.02 for ^{153}Tb; 12 for ^{154m}Tb (24.0 hr); 26 for ^{154g}Tb (24.0 hr); 0.0013 for ^{157}Tb; 0.001 for ^{158}Tb; 0.66 for ^{160}Tb; and 0.2 for ^{161}Tb (6.91 days) (the calculated values of the yields are analogous to the calculations of [5]). As we can see, when natural gadolinium is bombarded, the impurities of ^{156}Tb and other nuclides of terbium are extremely large.

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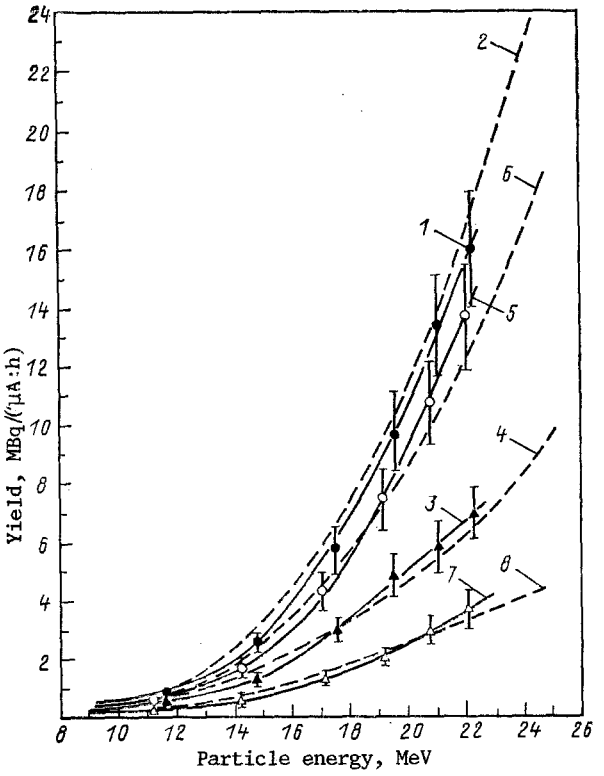

Fig. 1

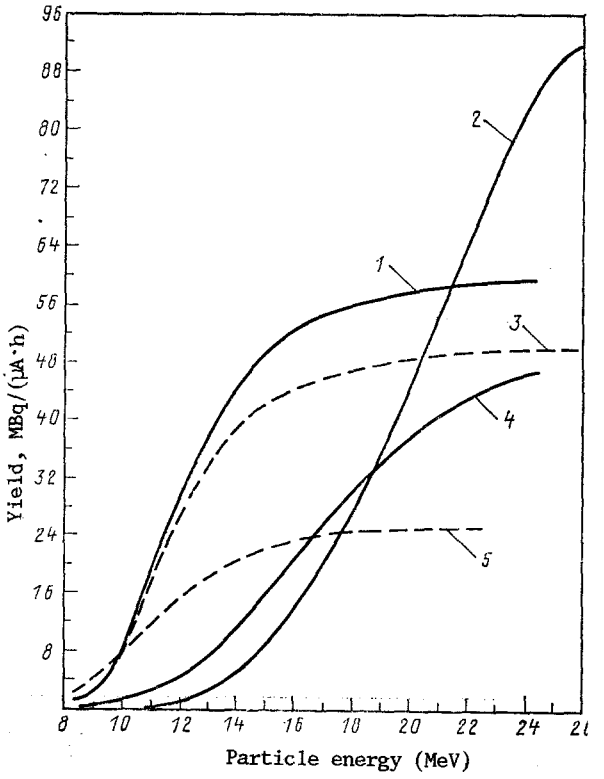

Fig. 2

Fig. 1. ^{155}Tb and ^{156}Tb yields versus proton and deuteron energies. The solid curves represent experimental values, and the dashed curves calculated values. For the (Gd + p) method: 1, 2) ^{155}Tb; 3,4) ^{155}Tb (reduced by a factor of 2). For the (Gd + d) method: 5, 6) ^{155}Tb; 7, 8) ^{156}Tb (reduced by a factor of 4).

Fig. 2. Yield of ^{155}Tb and of the impurity ^{156}Tb versus proton and deuteron energy for a thick target of ^{155}Gd and ^{156}Gd enriched to 98% (as found by using experimental and theoretical data) in the reactions: 1) ^{156}Gd(pn) ^{155}Tb (increased by a factor of 5); 2) ^{156}Gd(p2n) ^{155}Tb; 3) ^{156}Gd(pn) ^{156}Tb (increased by a factor of 5); 4) ^{155}Gd(d2n) ^{155}Tb; 5) ^{155}Gd(dn) ^{156}Tb (increased by a factor of 50).

The use of highly enriched isotopes of gadolinium (enrichment to 98-99%) and thin targets and the selection of optimal particle energy make possible a sharp decrease in impurities of other nuclides of terbium and a significant increase in the ^{155}Tb yield. Analysis shows that the following reactions are promising for producing ^{155}Tb for nuclear medicine from highly enriched isotopes: ^{155}Gd(pn), ^{156}Gd(p2n), and ^{155}Gd(d2n).

When the reaction ^{155}Gd(pn) ^{155}Tb is used, the reactions ^{155}Gd(p2n)^{154m,g}Tb and ^{155}Gd (p3n)^{153}Tb also take place. The ^{154m,g}Tb impurity can be eliminated by holding the target for about 8 to 10 days, and the ^{153}Tb can be eliminated by using protons having an energy less than or approximately 18.5 MeV [the energy threshold of the reaction (p, 3n) is 17.8 MeV].

The use of the reaction ^{155}Gd(p2n)^{155}Tb yields impurities via the reactions ^{156}Gd(pn) ^{156}Tb and ^{156}Gd(p3n)^{154m,g}Tb. The ^{156}Tb impurity can be reduced by using a thin target; the ^{154m,g}Tb impurity can be eliminated by holding or by using protons having an energy less than or approximately 20 MeV [the threshold of the (p, 3n) reaction is 19.3 MeV].

In the case of the reaction ^{155}Gd(d2n)^{155}Tb, the reactions ^{155}Gd(dn)^{156}Tb, ^{155}Gd(d3n) ^{154}Tb, and ^{155}Gd(d4n)^{153}Tb are responsible for impurity formation. Ways of reducing and eliminating the impurities are: for ^{156}Tb, using a thin target; for ^{154m,g}Tb, holding after bombardment; and for ^{153}Tb, using deuterons having an energy less than or approximately 21 MeV [the threshold of the (d, 4n) reaction is 20.2 MeV].

The values of the ^{155}Tb yield and the impurity content can be found from the yield curves shown in Fig. 2 for ^{155}Tb and ^{156}Tb, which are determined as follows. The theoretical curves of the ^{155}Tb and ^{156}Tb yields were calculated for the production methods (Gd + p) and (Gd + d). As we can see from Fig. 1, the theoretical and experimental curves of the yield match within the limits of the error of measurement. The theoretical curves in Fig. 1 were obtained by sum-

ming the theoretical curves of the yield for ^{155}Tb and ^{156}Tb, respectively, for the individual reactions (pn), (p2n), (p3n) and (dn), (d2n), (d3n). We assume that the fraction of the yield of each reaction in the total yield of radionuclide in Fig. 1 is the same for the calculated and experimental yields of ^{155}Tb and ^{156}Tb. For example, for the production method Gd + p→ ^{155}Tb at E_p = 22 MeV the reactions ^{155}Gd(pn)^{155}Tb, ^{156}Gd(p2n)^{155}Tb, and ^{157}Gd(p3n)^{155}Tb. According to Fig. 1, the total theoretical yield of ^{155}Tb at E_p = 22 MeV is 16.1 MBq/(μA·h). The theoretical yields of the individual reactions [in MBq/(μA·h) and their fractions in the total yield are 1.8 for (pn), 11%; 13 for (p2n), 81%; and 1.3 for (p3n), 8%. The total experimental yield of ^{155}Tb at E_p = 22 MeV is 15.2 MBq/(μA·h) (see Fig. 1). Then the reaction yields [MBq/(μA·h)] were: 15.2 × 0.11 = 1.7 for (pn), 15.2 × 0.81 = 12.3 for (p2n), and 15.2 × 0.08 = 1.2 for (p3n). Normalizing to these yields the relative behavior of the theoretical yield curves for these reactions, we obtain the yield curves for ^{155}Tb for a thick target in the reactions ^{155}Gd(pn), ^{156}Gd(p2n), and ^{157}Gd(p3n) by using the total experimental ^{155}Tb yield for the method (Gd + p). Analogous calculations also can be performed for the other yield curves in Fig. 1, and the yield curves of ^{155}Tb and ^{156}Tb can be obtained for the separate reactions (pxn) and (dxn) by using the total experimental yield.

In the yield curves obtained in this way for ^{155}Tb and ^{156}Tb for the individual reactions, the value of the yield was determined for natural nuclear targets. If an enriched isotope is used, this value increases significantly. For example, the yield 1.7 MBq/(μA·h) at E_p = 22 MeV is indicated for the reaction ^{155}Gd(pn)^{155}Tb. The ^{155}Gd content in natural gadolinium is 14.75%. When ^{155}Gd enriched to 98% was used, the ^{155}Tb yield increased by a factor of 98-14.75 = 6.64, to 1.7 × 6.64 = 11.3 MBq/(μA·h).

As we noted above, the reactions ^{155}Gd(pn), ^{156}Gd(p2n), and ^{155}Gd(d2n) are promising for the production of ^{155}Tb. For the last two reactions, a thin target should be used in order to reduce the ^{156}Tb impurity formed via the reactions ^{156}Gd(pn) and ^{155}Gd(dn). Figure 2 shows the yield curves of ^{155}Tb and ^{156}Tb of these five reactions for ^{155}Gd and ^{156}Gd nuclear targets enriched to 98%. The following conclusions may be drawn from Fig. 2 and the data presented above. When the reaction ^{155}Gd(pn)^{155}Tb is used, at E_p = 18.5 MeV the ^{155}Tb yield is 11.1 MBq/(μA·h); after holding for about 8 to 10 days, the actual ^{155}Tb yield is approximately 3-3.5 MBq/(μA·h). For 98%-enriched ^{155}Gd, the remaining 2% will contain some amount of ^{156}Gd and ^{157}Gd ($\leqslant$0.7%). The ^{156}Tb impurity formed via the reactions ^{156}Gd(pn) and ^{157}Gd(p2n) is $\leqslant$0,8% according to estimates. The impurities of other nuclides will be smaller by a large factor (as in the cases that we will examine below). In the reaction ^{156}Gd(p2n)^{155}Tb the ^{155}Tb yield at E_p = 20 MeV is 43 MBq/(μA·h) and the ^{156}Tb yield is 9.5 MBq/(μA·h), i.e., the ^{156}Tb impurity if 22%. For a thin target ΔE_p = 18-20 MeV, the ^{155}Tb yield is 17 MBq/(μA·h), and the ^{156}Tb impurity is approximately 3%. When 26-MeV protons and a thin target are used, ΔE_p = 20-26 MeV, and the ^{155}Tb yield is 48 MBq/(μA·h); after holding for about 8 to 10 days the actual ^{155}Tb yield is 12-16 MBq/(μA·), and the ^{156}Tb impurity is approximately 2.2%. In the case of the reaction ^{155}Gd(d2n)^{155}Tb, at E_d = 21 MeV the ^{155}Tb yield is 40 MBq/(μA·h), the actual yield after holding is 10-13 MBq/(μA·h), and the ^{156}Tb impurity is 1.2%. For a thin target ΔE_d= 17-21 MeV, the actual ^{155}Tb yield is 4-5 MBq/(μA·h), and the ^{156}Tb impurity is approximately 0.5%.

The examples considered here demonstrate the possibilities of the proposed methods of producing ^{155}Tb for nuclear medicine. Depending on the requirements for radioisotopic purity of ^{155}Tb, other production conditions also may be used (holding, target thickness, particle energy). We can see from the comparison that the proposed methods of producing ^{155}Tb are far superior to the production method using the reaction ^{153}Eu(α2n)^{155}Tb with respect to yield and radioisotopic purity.

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