

E. Radiopharmaceuticals

Photo-production of High Specific Activity Thulium-167 for Medical Use

K. SAKAMOTO, Y. SHIROTA,

Department of Chemistry, Faculty of Science,

A. ANDO,

School of Paramedicine,

K. HISADA

Department of Nuclear Medicine, School of Medicine

Kanazawa University, Kanazawa

Recent Studies have shown that rare earth radionuclides of high atomic numbers concentrate favorably in tumor tissues and bones (K. Hisada and A. Ando, *J. Nucl. Med.*, 14, 615 (1973), R.L. Hayes et al., *Proc. 21st Annual Meeting of Nuclear Medicine*, San Diego, June 1974, *J. Nucl. Med.*, 15, 501 (1974). Among these nuclides ^{167}Tm is reported to be one of acceptable tumor localizing agents in view of its decay characteristics, the half-life of 9.24 d and the EC decay followed by a 208 keV gamma-ray.

The present study concerns a new method of production of a high specific activity ^{167}Tm , via a reaction $^{168}\text{Yb}(\gamma, n)^{167}\text{Yb} \xrightarrow[17.7\text{m}]{\text{EC}} ^{167}\text{Tm}$. In a photo-reaction on ytterbium at moderate energies the 93.1 d ^{168}Tm is not produced and other long-lived nuclides ^{170}Tm ($T_{1/2}=130$ d) and ^{171}Tm ($T_{1/2}=1.91$ y) are produced in negligible amounts due to small cross sections of the (γ, p) reactions. Target materials used were a few tens grams of a commercial Yb_2O_3 of natural isotopic abundance and some tens mgr of a 22.5% enriched $^{168}\text{Yb}_2\text{O}_3$. Irradiation was performed in a

water pool just behind a 2 mm platinum converter with 30 MeV and 60 MeV electron beam from the 200 MeV Electron Linac of the Laboratory of Nuclear Science, Tohoku University. The ^{167}Tm yields were about 20 $\mu\text{Ci}/\text{gr}$ of natural Yb_2O_3 and about 50 $\mu\text{Ci}/10$ mgr of the enriched $^{168}\text{Yb}_2\text{O}_3$ in a 7 hr irradiation with an electron beam intensity of about 200 μA at 60 MeV, and an order of magnitude lower in a 30 MeV irradiation. The yield was dependent sensitively on the target position on the beam. Small photopeaks due to the 30.1 h ^{165}Tm (77.2 keV) and to the 7.7 hr ^{166}Tm (80.6 and 184.4 keV) were observed in gamma-ray spectra of the irradiated targets, which were less abundant in the 30 MeV irradiation compared with the 60 MeV irradiation. For the natural target, a chemical enrichment of Tm was tried by using YbSO_4 precipitation from an acetic acid solution in presence of Na-amalgam and a few mg of Gd hold-back carrier. Both the thulium rich fraction containing ~ 100 mg of the natural target material and the enriched $^{168}\text{Yb}_2\text{O}_3$ target were subjected to a cation-

exchange (Dowex-50Wx8, 100~200 mesh in a 1.5cm ϕ ×40cm column). The elution was performed at room temperature with 700~1,000ml of 0.12 M α -hydroxy-isobutylic acid of pH 3.8 at a flow rate of 15ml/hr. Fractions with an appreciable tailing of Yb were combined and subjected to the cation-exchange once again.

About 500 μ Ci of a high specific activity ^{167}Tm were prepared from a 22 hr irradiation of a 40 mg $^{168}\text{Yb}_2\text{O}_3$ with a electron beam of 200 μ A, and used for administration tests to Yoshida sarcoma-bearing rats and rats with fractured tibia (See A. Ando et al., Paper No. 137 of this Conference report).

Electrochemical Preparation of $^{99\text{m}}\text{Tc}$ -Phytate

I. IKEDA, O. INOUE, J. UCHIDA, Y. SUGISAWA, K. KURATA, S. KATO
Dainabot Radioisotope Laboratories, Ltd.,

We reported the eletrolysis method for labeling several kinds of compounds with $^{99\text{m}}\text{Tc}$ previously. As the results of further investigation, we found that there are some compounds such as penicillamin and phytic acid which can be easily labeled with $^{99\text{m}}\text{Tc}$ electrochemically.

The basic principle of electrochemical method is similar to that of electrolysis method on the reduction of $^{99\text{m}}\text{TcO}_4^-$ with stannous ion dissolved from the tin anode.

Pt-Sn plate was used for labeling, and stannous ion is released from tin plate by ionization tendency in electrolytic solution.

$^{99\text{m}}\text{Tc}$ was readily incorporated into phytic acid by this electrochemical method. Labeling efficiency of $^{99\text{m}}\text{Tc}$ -phytate averaged more than 95% under optimal condition. The reaction of $^{99\text{m}}\text{Tc}$ and phytic acid depends upon the pH of solution because the anodic reaction of tin in corrosion greatly depend upon the

pH of solution. The most suitable pH range for labeling is below 6 and $^{99\text{m}}\text{Tc}$ -phytate thus obtained precipitated with Ca^{2+} in pH above 4.

Though $^{99\text{m}}\text{Tc}$ -phytate is soluble in aqueous solution, it is accumulated into RES system by forming insoluble particles with Ca^{2+} in vivo. More than 90% of $^{99\text{m}}\text{Tc}$ -phytate was accumulated into liver in mice, and its biological half life in the liver was about 112 hr.

The stability of $^{99\text{m}}\text{Tc}$ -phytate was affected by various conditions. Though Pt-Sn plate stabilized the $^{99\text{m}}\text{Tc}$ -phytate solution, free $^{99\text{m}}\text{TcO}_4^-$ increased by removing the Pt-Sn plate from the solution. $^{99\text{m}}\text{TcO}_4^-$ was also released by increacing the temperature.

In respect that the electrochemical method does not need electricity and treatment of reductant, this might be one of the most convenient method for labeling of compounds with $^{99\text{m}}\text{Tc}$.