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IX. PRELIMINARY STUDY OF PRODUCTION ROUTES OF POSITRON EMITTING ISOTOPES OF MANGANESE

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INTRODUCTION

Positron emission tomography (PET) is usually associated with the short-lived radionuclides ^{11}C, ^{13}N, ^{15}O and ^{18}F which have half-lives between 2-110 minutes. However, many biochemical processes as well as in vivo transports of macromolecules have have typical times of many hours and days. In order to widen the application of PET to such processes we are investigating the production and use of more long-lived positron emitting nuclides.

The etiologies of neurological diseases like Parkinson's,Huntington's and Wilson's diseases, are still unknown and under investigation. One of the possible explanations is an abnormal andling of some trace elements, manganese among them. It is then of interest to produce positron emitting isotopes of manganese (^{51}Mn ($T_{1/2}$ = 46.2 minutes, β^+ abundance 97 %) and ^{52}Mn ($T_{1/2}$ = 5.6 days, β^+ abundance 27.9 %)) to be used in brain uptake study with PET. These isotopes can be

produced at PET-centres with low energy cyclotrons via $^{52}Cr(p,n)^{52}Mn$ and $^{54}Fe(p,a)^{51}Mn$ using targets with natural isotopic composition.

The aims of present work, are to estimate yields of the mentioned nuclides, and to elaborate technique for separationof manganese isotopes from chromium and iron targets.

MATERIALS AND METHODS.

A solid state detector (ultra-pure germanium) coupled to a multichannel analyzer was used for measurement of radioactivity.

Thick target yields estimation.

Three irradiations were carried out at the MC-17 cyclotron (Scanditronix, Sweden) at UUPC (Uppsala University PET-Centre, Sweden) for yields estimation of manganese isotopes.

A chromium metal target (thickness 0,45 g/cm^2, natural isotopic composition) was irradiated with 16.2 MeV protons to estimate yield of ^{52}Mn via $^{52}Cr(p,n)$ nuclear reaction.

A measurement was made 10 hours after end of bombardment (EOB) to allow for ^{52m}Mn decay. The thick target yield of 2 MBq/mAh (EOB).

A chromium metal target (thickness 0.3 g/cm , natural

isotopic composition) was irradiated with 8 MeV deuterons to estimate the yield of ^{51}Mn via ^{52}Cr(d,n) nuclear reaction. The only manganese isotope produced, was ^{51}Mn. Thick target yield was an order of 1 MBq/mAh (EOB). As an alternative route, a ^{54}Fe(p,α)^{51}Mn nuclear reaction was tested. For this purpose, an iron metal target (thickness 0.47 g/cm^2, natural isotopic composition) was irradiated with 16.2 MeV protons.

Taking into account the fact that ^{51}Mn has no convenient gamma-lines for radioactivity measurement, count rate at 511 keV peak was measured 1 hour and 15 hours after EOB. ^{56}Co was found the only positron emitting by-product. The difference between 511 keV count rate at different terms was accepted as ^{51}Mn activity. The thick target yield of ^{51}Mn was 63 MBq/mAh (EOB). No gamma emitting Mn isotopes were observed as impurities in this case.

Separation of Mn isotopes from Cr targets.

For the development of separation technique, chromium metal target (thickness 0.2 g/cm^2) was irradiated with protons at Gustaf Werner Cyclotron (The Svedberg Laboratory, Uppsala university). Proton energy range was from 36.8 to 34.5 MeV. In order to follow the separation process the gamma lines with energy 320 keV (^{51}Cr produced via ^{52}Cr(p,pn) nuclear reaction) and 744 keV (^{52}Mn produced via Cr(p,xn) nuclear reaction), were used. After the irradiation, the target was dissolved by heating in minimal amount of concentrated hydrochloric acid. Small aliquots (several tens microliters) of

the solution were taken for the development of separation technique.

Two different approaches (solvent extraction and ion-exchange) were tested.

Solvent extraction.

Solvent extraction using acetylacetone-chloroform (1:1) as organic phase and citrate buffer as water phase, was tested. At pH 5.6 of the water phase, this method provides about 75% yield of radiomanganese in two successive extraction. Manganese can be quantitatively reextracted with 6 M HCl. The disadvantages of this method are the necessity to adjust the pH value of the water phase between extractions, and poor stability of Cr^{3+} oxidation stage at the separation conditions which make radiochemist to carry out the separation very quickly. Ion-exchange separation. Two columns containing ion-exchange resins AG 50x8 and Dowex 1x8 respectively (1 ml each, 100-200 mesh) were equilibrated with 1M HCl.

The chromium (+3) ions were transferred into bichromate ions before ion-exchange. Aliquot containing chloride solution of chromium andmanganese isotopes was evaporated to dryness on the hot plate. Five ml of 5 M NaOH were added to dry rest and 5 ml of 30% H_2O_2 were carefully (drop by drop) added to the hot solution. The dry rest was dissolved with color changing from violet -green to yellow. Sodium hydroxide was neutralized with 4.3 ml of 6 M HCl and 10 ml 1M HCl were added to the

solution. At this stage solution changes its color to orange. The bichromate solution was loaded on the top of the Dowex 1x8 column. Chromium activity was quantitatively trapped at the column (it is clearly seen as an orange layer of resin) while Mn quantitatively passes through the column. The eluate was loaded on the top of AG 50 column in order to get rid of Na ions. Manganese activity is trapped with the efficiency of 95% and than can be eluted quantitatively with 5 ml of 6 M HCl. After evaporation to dryness radiomanganese is ready for dissolving in appropriate solution for further investigations.

RESULTS AND DISCUSSION.

The $^{52}Cr(p,n)^{52}Mn$ and $^{54}Fe(p,\alpha)^{51}Mn$ nuclear reactions were found acceptable for routine production of ^{52}Mn and ^{51}Mn, respectively. These reactions provide sufficient production rate even by use of targets with natural isotopic composition. Use of $^{52}Cr(d,n)^{51}Mn$ necessities an application of isotopicaly enriched target material which makes this way of ^{51}Mn production unpractical.

For the separation of radiomanganese from irradiated chromium targets technique using oxidation with following ion- exchange was chosen. The total separation time is in order of 1.5 hours, the yield-85-90%.

Work is in progress now to develop rapid and convenient method for radiomanganese separation from iron targets. The idea is to take away the bulk of iron via solvent extraction before

ion-exchange separation in order to minimize volumes of columns and solutions.

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