THE PRODUCTION OF SHORT-LIVED RADIOISOTOPES BY 14 MeV NEUTRONS

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Abstract

Because of the great importance of the use of radioisotopes of short half lives (several minutes to few hours) in medicine and in chemical and biological studies very rapid and simple methods of production of short lived carrier free ^{28}Al, ^{29}Al, ^{27}Mg, ^{24}Na and almost carrier free ^{34m}Cl, ^{78}Br, ^{80m}Br, ^{80}Br, and ^{128}I, using 14 MeV neutrons were developed. Similar methods can be applied for the production of other isotopes.

Introduction

The use of short-lived radioisotopes in medical, chemical and biological studies has the advantages of lower radiation doses and less hazards of radioactive contamination[1] . Besides, some elements have either short-lived radioisotopes or very-very long isotopes (e.g. Al or Cl).

Most of the radioisotopes are produced by nuclear reactors and cyclotrons[2] which are located only in large centers and thus short-lived radioisotopes are not readily available in places far from nuclear centers.

As there are many universities and research institutes which have 14 MeV neutron generators for teaching purposes, activation analysis, etc. and since it is quite possible that some hospitals will have in the near future neutron generators for neutron therapy it is very useful to develop methods to produce and separate short-lived radioisotopes using 14 MeV neutron generators.

Due to the relatively low neutron fluxes available by neutron generators (10^8-10^9 neutrons/sec cm^2) quite large samples (usually about 100 grams or more) have to be irradiated to obtain sufficient amount of radioactivity. As a result the irradiated material cannot be used without processing and the produced radioisotope has to be separated from the target. That this can be accomplished is either due to the (n,p) and (n,α) reactions which produce radionuclides of different elements other than the target and thus a chemical separation is possible or due to Szilard Chalmers processes for the (n,2n) and (n,γ) reactions[3] .

Preparation Processes

The separation processes should be rapid and simple in order to decrease the decay and to be of value also for small places. Appropriate processes are elution from a solid target on a column by an appropriate solution, mainly for the (n,p) and (n,α) reactions, or simple solvent extraction for liquid targets undergoing Szilard-Chalmer processes with the (n,2n) and (n,α) reactions.

(a) ^{28}Al (2.3 min) and ^{29}Al (6.52 min)[4] - Irradiation of 50 grams of Al_2O_3, in a glass column fitted with a sintered glass frit, with 2-3.10^8 n. $sec.^{-1}$ cm^{-2} 14 MeV neutrons for three minutes and elution with about 5 cc of 0.1 N HCl gives 40-80 μCi of ^{28}Al and about 0.5 μCi of ^{29}Al. The elution is carried out under vacuum by a rotary vacuum pump and the whole separation procedure takes less than one minute. Larger amounts of ^{29}Al can be obtained either by longer irradiation (up to a factor of 3) or mainly by using enriched $^{29}SiO_2$ (up to a factor of 20). The only contamination is small amounts of ^{27}Mg (∿ 0.1 μCi).

(b) ^{27}Mg (9.5 min)[5] - 50 grams of Al_2O_3 in a column are irradiated for ten minutes with a flux of 2-3.10^8 14 MeV $n.cm^{-2}.sec^{-1}$ and eluted with 5 cc of either pure water or saline solution. The eluted solution contains 15-25 μCi of ^{27}Mg with contamination of about 0.5 μCi of ^{24}Na. The elution carried out under vacuum takes less than one minute.

(c) ^{24}Na (15.05 hours)[6] - 500 grams of MgO are irradiated for twenty minutes and eluted with 200 cc of pure water. The eluted solution is evaporated to 2-3 cc. The whole procedure takes about 30-40 minutes and yields about 20-25 μCi of ^{24}Na.

(d) Chlorine - 34m (32.5 min)[7] - 250 ml of n-butyl chloride with 25 ml water are irradiated in a polyethylene separating funnel for 10 minutes. After irradiation the mixture of liquids is shaken thoroughly for one minute and left to be separated. The aqueous solution is separated into a small beaker and evaporated in a fumeboard to 2-3 cc which contains about 2 μCi of ^{34m}Cl and 10 μmole of Cl^- (formed by radialysis); the contamination with ^{38}Cl (37.3 min) is about 5%. The separation procedure takes 10 minutes. The reasons for choosing n-butyl chloride as a target are its density (desirable < 1 gram/cc) and its boiling point (desirable 70-80°C).

(e) Bromine - 78 (6.4 min), 80 (17.6 min) and 80m (4.4 hours)[7] - 250 cc of n-propyl bromide with 15 cc of water are irradiated for one minute, shaked and separated. The aqueous phase are evaporated to about 10 cc which contains about 70 μCi of 6.4 minutes ^{78}Br, 20 μCi of 17.6 minutes ^{80g}Br, 3μCi of 4.4 hours ^{80}Br and about 5 μmole of Br. The separation procedure takes about 6 minutes. In case the relative longer lived radioisotopes ^{80g}Br and ^{80}Br are required it is recommended to use 25-30 cc of water and to evaporate to 3-4 cc, besides irradiation for a longer period.

(f) ^{128}I (25 min)[7] - 250 ml of C_2H_5I with 25 ml are irradiated behind a paraffin sheet 2-2.5 thick for 10 minutes. After irradiation the mixture is shaked, separated and the aqueous phase is evaporated to 3-4 cc; the whole separation procedure takes about 10-12 minutes. The concentrated aqueous solution contains about 40 μCi of ^{128}I, 0.3 μCi of ^{126}I and 35 μmole of I^- anion.

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Recoveries

The recovery of the radionuclides by elution of the solid target material with a small volume solution (~5 cc) is about twenty per cent. Additional twenty to forty per cent can be eluted by using additional 20 - 25 cc of eluant, but it is not recommended usually for very short lived radionuclides because of the low specific activity obtained. Large volumes of eluant were used only for the preparation of ^{24}Na since its long half life (15 hours) gives time (~ 20 minutes) to concentrate the solution of the radionuclide by evaporation.

The recoveries from liquid - liquid extractions were about 30-40% for chlorine, 25% for bromine, where less water were used, and 40 - 50% for iodine. Higher yields of about 90 - 100% were obtained for radiochlorine, bromine and iodine when 0.01 gram I_2 or Br_2 was added to the organic liquid before irradiation and the elution was carried out with 0.1M sodium thiosulfate aqueous solution. The thiosulfate anion can be removed by an anion exchanger.

Health Hazards

Using very simple precautions of suitable shielding (few lead blocks) allows more than hundred preparations per week without exceeding the maximum permissible dose (100 millirem).

Costs

The target materials are used repeatedly and the only cost of material is that of the eluants which are inexpensive. The cost of irradiation is about $20-40 per an hour, not taking into account the capital cost of the neutron generator.

References

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