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Generators of alpha-emitting radionuclides 225Ac→221Fr→213Bi and 223Ra→219Rn→211Pb

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Generators of alpha-emitting radionuclides are of great interest for targeted therapy of cancer. Among possible generator pairs, 225Ac/213Bi and 223Ra/211Pb are the most promising. In addition to α-particle's properties attractive for nuclear medicine such as short range (50-100 μm) and high linear energy transfer (up to 100 keV/μm) in biological tissue, the mother radionuclides 225Ac and 223Ra have half-lives (9.9 and 11.4 days, respectively) convenient for production, transportation and medical use of the generators. A recently developed method of irradiation of natural thorium with medium-energy protons may provide Ci-amounts of 225Ac and 223Ra monthly [1] and promote large-scale applications.
In a usual generator scheme a mother radionuclide is adsorbed in a small volume at the top of chromatographic resin and a daughter is eluted. This scheme does not work well if used directly because the emitted α- and β-particles destroy the resin by radiation impact and radiolysis. The approach of generators 225Ac → 221Fr (4.9 min) → 213Bi (45.6 min) and 223Ra → 219Rn (4.0 s) → 211Pb (36.1 min) presented in this work consists in obtaining 213Bi and 211Pb via isolation of ultra short-lived intermediates 221Fr and 219Rn.
225Ac was recovered from a proton-irradiated thorium target and contained ~0.2% 227Ac (21.8 y) [2]. Actinium fraction was then adsorbed on a column filled with extraction-chromatographic Actinide Resin (TrisKem) using bis(2-ethylhexyl) methanediphosphonic acid as an extractant. 221Fr was eluted with HCl or HNO3 solutions of various (0.016-1 M) concentrations. Having passed the column, the eluate was pumped through a pipe long enough for 221Fr decay into 213Bi. 213Bi was concentrated on a second column also filled with Actinide Resin. The solution after the second column was directed to the first column forming a closed loop. As a result, 213Bi was accumulated in the second column up to secular equilibrium with 225Ac which was maintained till the moment of 213Bi elution. Circulation of eluate provided permanent removal of radiolysis products from the columns. In order to strip off 213Bi the second column was switched from the loop to an elution line and solution of 1 M HCl was passed. The total yield of ~90% was obtained. The radionuclidic purity of 213Bi solution was not less than 99.5%.
223Ra was recovered from the same thorium target and contained ~10% 224Ra (3.7 d). The latter generates a similar decay chain 224Ra → 220Rn (55.6 s) → 212Pb (10.6 h). A solution containing radium fraction was evaporated on a quartz support which was then heated at 800°C in argon flow. 219Rn and 220Rn were sublimed and blown out into a silicone pipe where they decayed. Generated isotopes of lead deposited inside the pipe. The values of argon flow, pipe length and accumulation time may be chosen to regulate the impurity of 212Pb in 211Pb down to 0.1-0.5%. After heating, 211Pb was stripped off from the pipe walls with small amount of 0.25 M HNO3 solution. The total yield of 80-85% 211Pb was obtained. The radionuclidic purity of 211Pb solution was better than 99.9% (except 212Pb). The presented generator based on gas chemical technique is highly resistant to radiation damage.

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