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Research Article

Guidelines for the preparation and isolation of Radionuclides produced with In-house Cyclotrons Bombardments

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Abstract

Carrier-free radioisotopes and cyclotrons are largely manufactured and sold in market in high prices, even many challenges are facing isotopes production in industry. Thus, we came here to introduce valuable and easy working conditions using different thick target materials under well-defined irradiations to separate some important isotopes. As a result, in carrier-free form, the present work has successfully isolated seventeen most common cosmogenic and biological radioisotopes recommended by the IAEA-TECDOC-1211, Charged Particle Cross-section Database for Medical Radioisotope Production: Diagnostic Radioisotopes and Monitor Reactions, and the IAEA, in addition to US National Nuclear Data Center (NNDC) databases.

Introduction

In November, 2005, the United States Atomic Energy Commission (USAEC) has launched a novel programme [1,2], to enhance the needs for developing *cyclotron*-generated and reactor-*produced radionuclides* (i.e. Tc-99 m, I-123, I-124, Zr-89, Cu-64, Ga-67, Ga-68, In-111, Y-86 and Sc-44) (or called as *reactor-produced radioactive isotopes (RPRI); energy range 3 MeV-20 MeV*) distribution including their marketing process especially for medical applications [3,4]. In 5yrs, the programme was being in process, which perceived the work on radiopharmaceuticals. *Cyclotron producing radioisotopes* had been largely expanded especially in clinical application [3]. Even though, as noted by USAEC, it was not marketed as the nuclear *RPRI*, which nowadays plays a foremost active role in scientific research, and recent developments in several arenas especially "molecular biology" [5,6]. The challenge that brakes the distribution of this important technique attributes to the imbalance between the expenditures and the gained products, where to produce a little quantity of radioactivity by the expensive charged particle accelerator (CPA), this entails high expenses. Taking into consideration this challenge, the cyclotron produced carrier free radioisotopes (CFSR) can overcome these obstacles and presents its assets as desirable technique in major fields [5].

A carrier free isotope or CFSR has been considered the only technique that needs no quantifiable masses of stable isotopic elements of the same component present [7]. Through charged particle radioactivity, the carrier free isotopic elements are generated where their atomic numbers (Z) (or the chemical identity) are different from Z-values of the original target substance/material (TM) [8]. With the usage of distinct chemical separation systems, such as solvent extraction and/or ion exchange chromatography (IEC), macro extent of TM can be separated from the tiny quantity of radioactive material then decontaminated and organised as per CFSR [9,10]. CFSR may be implemented in several ways. These could be inoculated into humans and animals due to the insignificant chemical tolerance, i.e. As^{74} [11]. These could be utilised for radiochemical investigations in very little contents, (<0.01µmolar). They can also be implemented for preparing of insubstantial lightweight-counting standards, abolishing the amendments aimed at scattering and self-absorption correction [12].

Basic principles of CPAs yield radioisotopes that generally are neutron-deficient nuclei regarding the nuclear-powered

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stable line, and that falling-off through positron discharge or even electron seizure [13,14].

RPRI designed by neutron capture are too much neutron, falling-off thru β-particle emission [15]. For a particular element, the rate of radioisotopes' half-lives is commonly dissimilar. Therefore, by various applications, the selection of radioisotopes of a certain element, of varied properties of radioactivity is accessible. As an example, the shorter-lived isotopic nuclide of 65-d Sr^{85}, flexible to an appropriate system of scintillation counting feature, is utilised with noxious fission isotopic product (~30-yr Sr^{90}), that only decays via β-particles radiation (β^- and β^+). So that, Sr^{90} decays (100% of the time with $E_{\beta,\,max}$ = 546 *KeV*) to Y^{90} then the later decays to the stable isotope Zr^{90} (~64-hr) by also β-emission. Hence, after approximately 7x64 hrs (18.67 d), the Sr^{90} and Y^{90} are reached secular equilibrium and decay at the same rate. The longer-lived radionuclide of 2.6-yrs Na^{22} or even 105-d Y^{88} can be utilised in operations implement certain isotopes of awfully short half-lives as 64-hr Y^{90} or even 15-hr Na^{24}. Cyclotron irradiation may frequently generate radionuclides with no association of other radioactive isotopes of an identical element, that in other way is only gained by the reactor irradiation of electromagnetically enriched isotopes [16]. Electromagnetic source equipment for this type of separation is usually made up of functional subunits, so arranged that any component that fails in service can be removed and replaced by remote control without the necessity for complete chemical and mechanical servicing of the entire unit. It has also been suggested that the receiver be designed so that individual collectors can be readily handled through recovery chemistry. Remote handling of all systems behind suitable barriers appears to be practical. Wash chemistry and recovery chemistry would be handled in areas isolated from operational and servicing areas [17]. As an illustration, 3-yr Fe^{55} may arranged with no 45-d Fe^{59}. Lastly, the fraction of elements which does not hold practical radioisotopes is completed with nuclear reactor irradiation. They comprise Al, Ti, and V. Exclusively, cyclotron irradiations have been designed at developing ~10^6-yr Al^{26}, 3.08-hr Ti^{45} and 16.2-d V^{48}.

The commencement of a database for the invention and circulation of cyclotron fashioned CFSR asked for the growth of a number of actions which had not been beforehand described including the overviews of the amendments into published trials [11,18] and which were not utterly acceptable for the envisioned practice as of short chemical yield or a deficient product purity. Moreover, the fairly scant data of cyclotron have also posed another challenge for the current study.

Materials and Methods

The chemical procedures were operated in Pyrex glassware excluding sodium isotope (Na^{22}) and when employing hydrofluoric acid (HF). However, to get minimal exchange medium of Na^{22} along with the sodium exists in the silicate glass, Vycor containers (96% SiO_2 and 4% B_2O_3) were utilised. Polyethylene (PE, $(C_2H_4)_n$) and alternatively HF-inert materials have been implemented to capacitate HF solutions.

The measures are designed for radiochemical separations of such elements wherefore artificial-shaped radionuclides have been detected. Yet, the realised separations encompass the supplementation of labile isotopic ratios of matching element as carriers, has achieved reference chemical procedures with sub-µg amounts of radioactive classes pursued. Meanwhile, carrier-free radioisotopes can be favourite products and those techniques of separation are commonly unfitting. The separation measures for the sub-µg extents of isotopes ought to be considered with the available instruments which are typically mass-independent, alike to coprecipitation along with solvent extraction, IEC, and non-isotopic carriers. The actions, given in this paper, were settled from these practices. In addition, every so often, hypothetical yield assessment is the only information offered. A comparison study has been carried out with previous published works, which were utilised as virtual references in scheduling irradiations. With 8MeV protons, deuterons of 15MeV energy, and with protons of 15 and 20MeV energies, the radioactivity for the manufacture of the CFSR has been investigated in the current article.

Via ɣ-radiation spectroscopy, the analysis (apart from W^{181}, Cd^{109}, and Fe^{55}) of the isolated activity equipped with a well-formed thallium (Tl)-doped sodium iodide (NaI) modulated with a scintillator system armed with single-channel pulse-height analyser (SCPHA) has been implemented (linearity within 1% of deviation for radiation energy; stability within 1%/24hrs of fluctuation; temperature stability of channel position and width ± 0.002%/°C; deadtime 23 µsec.; resolution 0.08 µsec.) and the spectra taken with SCPHA were in a good agreement with these taken by cathode ray tube (CRTA) analyser (both analyser window widths were set at approximately 500 mV). At one time, the SCPHA used for a pulse height analysis has a lower and an upper level discriminator, and produces an output logic pulse when an input pulse lies between the discriminator levels. All pulses in a specific range are routed to a scalar (counter). The SCPHA was set to a narrow ΔV range and then the lower level discriminator has been swept through a series of voltages using an SCPHA Sweep device. All pulses outside the SCPHA window were lost, so that the smaller the window for better resolution, the longer the time required to get adequate counts. In this experiment, it was not necessary to use multi-channel pulse-height analyser (MCPHA) even it would be suggested to collect the entire spectra at once because SCPHA is a sufficient analyser used in establishing coincidence between nuclear decays which is better than most MCPHAs that are busy themselves in digitizing many uninteresting events increasing the dead time. Besides, SCPHA is much cheaper and simpler in design than MCPHA.

However, for W^{181}, Cd^{109}, and Fe^{55}, which do not possess distinguished energetic ɣ-rays, end-window variable-geometry proportional flow counters have been utilised. To assess counter effectiveness for the miscellaneous radiations of interest, standard specimens of various isotopes were brought from either the National Bureau of Standards (NBS) or checked analytically with the assistance of external quality control (QC) with other testing laboratories. Considering the unavailability of the standards for specific isotopes, the emerging faced problem in this research, counter efficacies were assessed by adopting a method of interpolation among the obtainable standards. For specific situations of very tiny radiation energy density, that was exigent to roughly estimate the counter effectiveness regarding the physical ordering and assets of the counter electronic parts.

On the whole, analyses can be performed not as much of 10% absolute uncertainty (AU) and the latter term is usually emerged from serious sources related to the thick target yield data arise from such places of origin.

Results and Discussion

In the current programme, the achieved irradiations have provided seventeen dense target yields of radioisotopes (n = 3) (Table 1). We also presented in this table, the incident elements used in irradiation, the target chemical, the yield-dense target adjusted for falls experienced in the chemical operation and falling-off from the time of termination of the irradiation to analysis time. Besides, we extracted the yield-dense target numbers published in the previous studies for matching radioisotope, in addition to the main causes of uncertainties in the yield-dense targets. By hand, the chemical operating of the exposed targets, period and intensity of irradiations, in addition to the radiometric assays of the isolated carrier-free activities have been served to assess the thick target yield data. The yields of activities after chemical isolation and the combined beam streams of the irradiations have been indicated by experimental runs.

In general, the absolute yields reached (≥90%) and the assessed errors were allotted (≤5%.) In limited instances, the yield was hard to measure and the possible absolute error reached higher than 30%. The short-lived isotopic elements were needed for collating the total yield information and the rapid separation. For the aggregated present data, we allotted the probable absolute errors of the order of 5-10%. Under unsealed irradiated settings, the target evaporation was regarded as a probable cause of non-reproducibility of cyclotron-produced of irradiation yields. This was confirmed with Sr^{85} yield, where the irradiations made with rubidium chloride (RbCl) were greater than the anticipated variations. Further radioisotopes of the matching element were formed for certain itemised radioisotopes. Those cases had complicated the analysis, where the segregation of the wanted isotope might not be realised by chemical means. For instance, two bismuth (Bi) radioactive isotopes were interfered with the radiometric analysis of Bi^{207}.

To permit an adequate decay of 6-d Mn^{52} isotope for an Mn^{54} accurate assay, an extensive delay period was necessitated. However, these forms of decaying systems along with the involvement of half-lives have increased the uncertainty levels and added some erratic yield determinations. Based on the routine analyses, the chemical isolation methods have successfully been performed and their results are discussed below. But actually, those methods were based on certain procedures used suitable techniques for CFSR as coprecipitation (CPT) along with IEC, non-isotopic carrier, or solvent extraction, or could be an integration of them.

We had registered an incomplete decontamination and some meagre yields as results of great deviations from the specified conditions, actually attributed to the reagent concentrations and volumes. Even though, the produced isotopes attained were ostensibly carrier-free, e.g. no non-energetic atoms are purposefully supplemented, roughly inactivated substance remains unvaryingly existing. The origins of the uncleanness are attributed to the reagents employed in the segregation and the contaminations in TM. Therefore, we suggest careful selection of reagents used and active purification of the TM, in order to practically get carrier-free products.

Summary-perspectives in tentative nuclear experiences

From our successful experience in this research, we suggest the following practices for carrier-free separation for radioisotopes (CFSR), depending on the type of isotope.

I. Be^7. TM: lithium (Li).

Product (Prod.): 99 + %Be^7, in hydrochloride acid (HCl) or nitric acid (HNO_3).

Production reactions (Prod. Reac.): Li^7 (p,n) Be^7 or Li^7 (d,2n) Be^7.

Other important reactions (OIR): None.

Chemical separation principles (Chem. Sep. Princ.): Be^7 is persisted on ferric hydroxide ($Fe(OH)_3$). After that, Iron (Fe^{III}) is detached by IEC.

Recovery (Rec.) obtained (98%); Time required (TR): nearly 4 hrs.

The Li metal, about 10g, is etched from its Cu with the assistance of 0.02N HCl. After acidification of the ensuing solution with HCl, any precipitates exist are separated by filtration. Then, 30mg of Fe^{III} carrier are supplemented, and the solution is just kept ammoniac. After 1hr of digestion, the $Fe(OH)_3$ has been segregated by separation through Whatman™ cellulose paper (Grade 42). Be^7 activity has quantifiably reserved in the solid phase. The latter phase is desolated in a minimum volume of HCl and then reprecipitated with ammonium hydroxide (NH_4OH). After 1hr digestion, the precipitate has isolated by centrifuge and washed two-times with water, and re-desolated in 5ml (12N HCl).

II. The anion exchange resin Amberlite IRA-400 is proved as an effective and reusable catalyst, thus, this column has been prepared and conditioned with 12N HCl. The solution is gone across this column, the Fe being retained and the Be^7 transverses over. The column is rinsed thrice using 12N HCl. To break down the organic matter (OM), the whole eluate has evaporated to near dryness and dealt with HNO_3 solution. As if wanted, Be^7 is transformed to chloride form by recurrent evaporation with $HClNa^{22}$. TM: magnesium (Mg).

Prod.: 99 + %Na^{22}, in HCl or HNO_3 solution.

Prod. Reac.: Mg^{24} (d,α) Na^{22}.

OIR: Mg^{25} (d,n) Al^{26}; Mg^{26} (d,2n) Al^{26}.

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Table 1: Yields of dense cyclotron marked-targets.

Isotope	Target	Incident particle	Rate of production mC/mA-hr			Major sources of uncertainties# (This work)
			This work	Other works in the literature		
Be^7	Li	15MeV deuterons	48±10	Low	[19]	a,e
		19MeV deuterons		2.00±25%	[20]	
		8MeV protons	77±12	170		
		20MeV protons	166±25			c,f
Na^{22}	Mg	14MeV deuterons	2.00±0.15	1.00±≥25%	[21]	e,f
		15MeV deuterons		1.75	[22]	
		20MeV protons		0.40		
V^{48}	Cr	15MeV deuterons	75±25	Low	[23]	b,c
	Ti	15MeV deuterons				
	TiO_2	15MeV deuterons	125±12			c
Mn^{52}	Cr	15MeV protons	144±20	3.00	[24]	b
		19MeV deuterons	4.50±2.00	8.00±≥25%	[25]	a,b
		8MeV protons			[24]	
		20MeV protons		80		
Mn^{54}	Fe	14MeV deuterons	1.40±0.30		[26]	d,e
		15MeV deuterons				
	Cr	20MeV protons	0.50±0.20	0.50	[27]	b
	$80\%Cr^{54}$	20MeV protons	87±10			a
Fe^{55}	Mn	15MeV deuterons	3.70±0.70	0.67	[28]	a
		19MeV deuterons		0.02±≥25%		
		20MeV protons	13.5±5.00	10	[29]	
Co^{57}	Fe	15MeV deuterons	9.50±2.00	5.00±≥25%	[30]	a,d
		19MeV deuterons				
	Ni	20MeV protons	22±6.00	50	[31]	a
Zn^{65}	Cu	14MeV deuterons	15.0±1.50	0.50±≥25%	[32]	a
		15MeV deuterons		3.50		
		20MeV protons		9.30		
Ga^{67}	Zn	15MeV deuterons	85±20		[33]	a,c
As^{74}	Ge	15MeV deuterons	31±10		[34]	a,b,c
		19MeV deuterons		10.0±25%		
		15MeV protons	170±50	50		a,b,c
Sr^{85}	RbCl	15MeV deuterons	16.0±6.0	0.13±25%	[35]	c,f
		19MeV deuterons		0.60±25%		e
		15MeV protons	23.0±4.0			
Y^{88}	SrO	14MeV deuterons		0.10/±25%	[36]	
		15MeV deuterons		38		
		19MeV deuterons		1.00±25%		
		15MeV protons	32±3.0	20		a,e
Cd^{109}	Ag	15MeV deuterons	2.60±1.0	2.00±≥25%	[37]	a,b
		19MeV deuterons				
In^{111}	Cd	15MeV deuterons	18.0±12.0		[38]	a,c,d
Ce^{139}	La_2O_3	15MeV deuterons	3.10±1.60		[39]	a, c
		15MeV protons	5.70±2.90			
W^{181}	Ta	15MeV deuterons	4.50±2.00	0.0005±≥25%	[40]	a,b,c
		19MeV deuterons				
Bi^{207}	Pb	20MeV protons	0.80±0.50		[41]	a,d,f

Key origins of uncertainty:
a - insignificant number of irradiations completed.
b – uncertainty in the yield.
c - radiometric assay in doubt.
d - different radionuclides restricted the radiometric test.
e - beam current uncertain.
f- possible target evaporation.

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Chem. Sep. Princ.: Al^{26} is taken off by carrying on beryllium hydroxide ($Be(OH)_2$). The majority of Mg is removed through the precipitation with ammonia (NH_3). Remaining Mg has done away with IEC.

Rec.: 96%; TR: ~ 12hrs.

The Mg cup target is floated on 50ml of water in a Vycor beaker. Minor volumes of HCl are supplied pending an activity utterly abolished. Approximately 10g of Mg are desolated, 25ml of HCl added and the solution watered down to 200ml. Sequentially, precipitates are separated by filtration.

To regain Al^{26}, 25mg of Be carrier are added then with NH_4OH the solution fetched to a methyl red endpoint. After cooling to room temperature, the Beryllium hydroxide ($Be(OH)_2$) is detached by centrifugation, washed two-times with water, and kept back to recover Al^{26}.

The supernatant solution is condensed by the evaporation to 100ml and saturated with NH_3 gas in ice bath. Magnesium hydroxide ($Mg(OH)_2$) precipitate is separated by filtration, with suction across a fritted glass funnel. The precipitate is suspended in the funnel thrice with NH_4OH and dehydrated by suction every once. The washes and filtrate are mixed and evaporated to near dryness, 25ml HNO_3 supplemented, and the solution is evaporated to near dryness. The prescribed action is repeated twofold to get rid of ammonium chloride (NH_4Cl) extant. 25ml water is added to the deposit of the last evaporation. If unsolvable material is existed, using filtration, we disposed it.

The sulfonic acid ion-exchange resin (Dowex-50) column is conditioned with 2N NH_4OH until the effluent reaches a pH of 9. At a rate of about $0.3ml.min^{-1}.cm^{-2}$, the solution is passed over the column. The column is washed with 200ml of water then with 0.35N HCl, the Na^{22} is eluted. In a Vycor beaker, the eluate is evaporated near to dryness and NH_4Cl is yet once more eliminated with HNO_3. Furthermore, with the later acid the OM is further vanished and if wanted, Na^{22} can be prepared carrier free in HCl solution by recurrent evaporations using HCl.

For especial situations, when inactive Na is recognised in existence with Mg target, or where a definite activity of the command of 10c/g is satisfactory, the practice could be condensed to a certain degree. If 1mg of Na is existing, the overall purification step could be affected by the precipitation of sodium magnesium uranyl acetate. Uranium (U) is precipitated with NH_4OH and the remaining Mg is then getting rid of by CEC as outlined above. Al^{26} can be recovered by one of the following methods [42,43].

III. V^{48}. TM: TiO_2.

Prod.: 98 + %V^{48} + V^{49} in HNO_3 or HCl solution.

Prod. Reac.: Ti^{47} (d,n) V^{48}; Ti^{48} (d,2n) V^{48}; Ti^{48} (d,n) V^{49}; Ti^{49} (d,2n) V^{49}.

OIR: Ti^{48} (d,α,) Sc^{44}; Ti^{48} (d,α,) Sc^{46}; Ti^{49} (d,α,) Sc^{47}; Ti^{48} (d,αp) Ca^{45}; Ti^{46} (d,n) V^{47}; Ti^{47} (d,2n) V^{47}.

Chem. Sep. Princ.: A sodium carbonate (Na_2CO_3) – sodium nitrate ($NaNO_3$) fusion of the dissolute target leads to unsolvable titanium dioxide (TiO_2) and a solvable vanadate material (i.e. trioxovanadate VO_3^-, tetraoxovanadate VO_4^-, decavanadate $V_{10}O_{28}^{-6}$). Calcium (Ca) and scandium (Sc) are co-deposited with ferrous hydroxide ($Fe(OH)_2$) from high concentration of caustic NaOH solution. V in a form of vanadyl ion or oxovanadium(IV) cation, VO^{2+}, is persisted in $Fe(OH)_3$ along with adjusting pH conditions. Fe is eliminated via IEC.

Rec.: ~90%; TR: ~6 hrs.

To a fine powder, the target, nearly 5g of TiO_2 is pulverised and deposited in a Ni-crucible. 0.2g $NaNO_3$ and 10g Na_2CO_3 are supplemented and the solids are blended in a thorough manner.

For 45min and at 700°C, the blend is fused. The melt is cooled off, pulverised, and drained away with cold water to secure complete V-dissolution. Using evaporation technique, the bulk of the solution is condensed to 100ml then lomg Fe^{III} carrier is supplemented, a bonus amount of NaOH has been added to ensure $Fe(OH)_3$ precipitation. The deposition has performed again and served to get away both Sc and Ca. After $Fe(OH)_3$ removal by centrifugation, the solution is acidulated and heated to thrust carbon dioxide (CO_2) out. 10g of Fe^{III} carrier are added and the pH is cautiously adjusted to 6.2 with NH_4OH. After 30min period of digestion, $Fe(OH)_3$ is segregated by centrifugation. As vanadyl ion, apparently V is quantitatively reserved by the precipitate, so that, the precipitation is performed more than two times. Besides, in a minimum quantity of 4N HCl, the ending deposition is dissolute and the obtained solution is transverse across an Amberlite IRA-400 column that with 4N HCl is being conditioned.

To eliminate the remaining V-activity from the column, an 4N HCl wash-down has been completed. The eluate is evaporated near to dryness, and dealt in sequence with HNO_3 to get rid of NH_4Cl and to break down OM. By evaporation with HCl, the Cl^- solution is resulted. But it would also be perceived that this process does not eliminate aluminium. Besides, the target backing or the container must be Ni.

IV. Mn^{52} and Mn^{54}. TM: Cr.

Prod.: 98+% Mn^{52}+ Mn^{54}, in HCl or HNO_3 solution.

Prod. Reac.: A. deuteron bombardment, Cr^{52} (d,2n) Mn^{52}; Cr^{53}(d,n) Mn^{54}; Cr^{54} (d,2n) Mn^{54}.

B. proton bombardment, Cr^{52} (p,n) Mn^{52}; Cr^{53} (p,2n) Mn^{52} ; Cr^{54} (p,n) Mn^{54}.

OIR: A. deuteron bombardment, Cr^{50} (d,α) V^{48}.

B. proton bombardment, Cr^{50} (p,2p) V^{49}.

Chem. Sep. Princ.: Mn^{52} and Mn^{54} are carried on $Fe(OH)_3$ from NaOH-Br_2 solution.

Cr and V are reserved in the liquid-supernatant layer. To separate from the residual Na, Mn^{52} and Mn^{54} are carried on $Fe(OH)_3$ from NH_4OH, and Fe is separated by IEC.

Rec.: ~95%; TR: ~6hrs.

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With HCl, the Cr is etched from its Cu-backing and 10-15g are solvated. 20mg Fe^{III} are mixed and the solution is evaporated near to dryness. Then, the residue has been solvated in a min. HCl, whereby the volume is adjusted to 25ml. The solution is decanted, blended with cold 7N NaOH (300ml) saturated with liquid Br_2. The ensuing $Fe(OH)_3$ precipitate has quantitatively carried Mn, whereas Cr and V are reserved in the supernate.

On a hot-plate and after 30min of digestion, the precipitate with suction is separated by filtration through a fritted-glass funnel. The precipitate is watered until no colour is distinguished in the washing water, which then solvated in a min. 1N HCl. The deposition (by means of a usage of NaOH-Br_2 solution (150ml)), filtration, and washing are then reiterated thrice. The last deposition substance is solvated in a minimum quantity of HCl, then a surplus volume of NH_4OH is added to deposit $Fe(OH)_3$. The precipitation is repeated two-times to isolate the activity from residual Na and the ending precipitate is solvated in 4N HCl (5ml).

With 4N HCl, the Amberlite IRA-400 column is conditioned, the solution is passed across the column, and the activity is washed through with 4N HCl. Fe is retained by the column. The elute is evaporated near to dryness and dealt with HNO_3 in order to eliminate OM. If wanted, the Mn^{52} may be obtained in HCl solution by processing a repetitive evaporation with HCl [42,43].

V. Mn^{54} and Co^{56}, Co^{57}, Co^{58}. TM: Fe.

Prod.: 98 + %Mn^{54} and 98 + % pure aggregated Co activity in HCl or HNO_3 solution.

Prod. Reac.: Fe^{56} (d,α) Mn^{54}; Fe^{56} (d,2n) Co^{58}; Fe^{56} (d,n) Co^{57}; Fe^{57} (d,2n) Co^{57}; Fe^{57} (d,n) Co^{58}; Fe^{58} (d,2n) Co^{58}.

OIR: Fe^{54} (d,α) Mn^{52}.

Chem. Sep. Princ.: Using ethyl ether ($(C_2H_5)_2O$), Fe has been extracted. Mn and Co are discriminatorily eluted from an anion exchanger.

Rec.: 98%; TR: ~8hrs.

With HCl, the Fe target is etched until basically all the activity is in solution. About 10g are dissolved. 20mg of cold liquid Br_2 are supplied to oxidise the existed Fe to Fe^{III}. The solution is permitted to stand for 2hrs and after that has evaporated to 40ml. Fe is extracted with $(C_2H_5)_2O$ and with 3- extractions more than 95% of Fe has been removed. The solution is evaporated to almost dryness then 8N HCl (5ml) are added. Using the later acid with the same concentration, Dowex-1 column is conditioned. The solution is passed over the column and by using 8N HCl, the Mn has eluted. 10mg fractions are collated, then checked for activity. Once the Mn is being entirely eluted, the eluant is changed to 4N HCl to eliminate the Co. Similarly, 20mg portions are collated and checked. Fe abides on the column. Mutually, the Mn and Co portions are dealt with HNO_3 to eliminate OM. What's more, the activity is transformed to the chloride by reiterating the evaporation action with HCl.

VI. Fe^{55}. TM: Mn.

Prod.: 99 + % Fe^{55}, in HCl or HNO_3 solution.

Prod. Reac.: Mn^{55} (p,n) Fe^{55}; or Mn^{55} (p,2n) Fe^{55}.

OIR: none.

Chem. Sep. Princ.: Mn is separated by IEC.

Rec.: 99%. TR: ~4hrs.

From its backing, the Mn is etched using HCl. Approximately, 10g are solvated. The resultant solution is condensed by drying to 20ml, then with water has diluted to 40ml. With 4N HCl, the Dowex-1 column is conditioned and then the solution passed across the column. Moreover, Fe is retained and Mn has eluted from the column with 4N HCl. Subsequently, the Fe^{55} has eluted with HCl (0.5N). Near to dryness, the eluate is dried and dealt with HNO_3 to eliminate OM. Notably, the HCl solution is collected by repetitive evaporation of HCl.

VII. Zn^{65}. TM: Cu.

Prod.: 99 + %Zn^{65}, in HCl or HNO_3 solution.

Prod. Reac.: Cu^{65} (d,2n) Zn^{65}; or Cu^{65} (p,n) Zn^{65}.

OIR: Cu^{65} (d,α) Ni^{63}; Cu^{65} (d, αp) Co^{60}.

Chem. Sep. Princ.: By an anion exchanger in a weak HCl, Zn^{65} is reserved. While the other divalent transition dements are not.

Rec.: 98%; TR: ~5hrs.

VIII. By 1N HNO_3, the activity is etched from the Cu-target. An amount between 5 and 10g are solvated. The ending solution is dewatered to dryness and transformed to chloride by proper evaporation with HCl. The residue has been solvated using a min. 1N HCl that after a full evaporation to dryness. A Dowex-1 column is arranged and with 1N HCl has conditioned. The solution is passed over the column, which is then washed with 1N HCl till Cu cannot be distinguished with H_2S addition to the effluent. Co and Ni are not reserved, however then, with 0.5mN HCl, the Zn is eluted. To almost dryness, the solution is dried out and dealt with HNO_3 to eliminate OM. A chloride solution may be gained with HCl evaporations.Ga^{67}. TM: Zn.

Prod.: 98 + %Ga^{67}, pure aggregated Ga activities in HCl or HNO_3 solution.

Prod. Reac.: Zn^{66} (d,n) Ga^{67}; Zn^{67} (d,2n) Ga^{67}.

OIR: Zn^{66} (d,2n) Ga^{66}; Zn^{67} (d,n) Ga^{68}; Zn^{68} (d,2n) Ga^{68}; Zn^{66} (d,α) Cu^{64}.

Chem. Sep. Princ.: Ga is carried on $Fe(OH)_3$ from NH_4OH solution. Cu is retained in the supernate. Fe is isolated by precipitation from high concentration of KOH solution. K and the remaining Cu are sorted out by IEC.

Rec.: 80 + %; TR: ~6hrs.

Citation: Aljerf L, Dastan D, Sajjadifar S, Bhatnagar S, Ukaogo PO, et al. (2019) Guidelines for the preparation and isolation of Radionuclides produced with In-house Cyclotrons Bombardments Open Journal of Chemistry 5(1): 020-029. DOI: http://dx.doi.org/10.17352/ojc.000013

About 5g of the Zn target is dissolved in a min. quantity of HCl and the solution is diluted to 100ml. 20mg each of Fe^{III} and Cu^{II} carriers are added. The pH is cautiously maintained at 5.5 with NH_4OH and the solution is permitted to digest for 15min. The precipitate is getting rid of by centrifugation then solvated in HCl. The 2nd precipitation is executed to guarantee a proper isolation from Cu. In a minimum of HCl, the last precipitate is dissolved, the volumetric content is made up at 20ml with water. At that point, an additional quantity of KOH is supplemented. The precipitate is segregated by centrifugation after heating to the boiling point (b_p), and the supernate containing the activity as gallate ion ($C_7H_5O_5^-$) is maintained. The precipitate is solvated in HCl and a 2nd precipitation is carried out. The supernates are intermingled, neutralised through HCl addition, then a surplus amount of HCl is added to give a solution 3N in HCl. The solution has gone across a Dowex-1 column and is conditioned with 3N HCl. 3- column volumes of 3N HCl are moved across the column to eliminate K and the remaining Cu. With a 0.2N HCl, the Ga has been eluted. OM is eliminated by evaporation with HNO_3 after heating the solution to the desired degree near to dryness. By evaporating the solution with HCl, the conversion to chloride is affected.

IX. As^{73}, As^{74}. TM: Ge.

Prod.: 98 + %As^{73} + As^{74}, in HCl-Cl_2 solution.

Prod. Reac.: Ge^{72} (d,n) As^{73}; Ge^{73} (d,2n) As^{73}; Ge^{73} (d,n) As^{74}; Ge^{74} (d,2n) As^{74}.

OIR: Ge^{70} (d,n) As^{71} Ge^{71}.

Chem. Sep. Princ.: In the existence of Cl_2, Ge is separated from HCl by distillation process. Then, As is distilled out from an HCl-CuCl medium.

Rec.: 90 + %; TR: ~6hrs.

Approximately 5g of the Ge target is aged for 10-15d to let As^{71} decaying which is sequentially solvated in aqua regia (Royal water, HNO_3 + 3 HCl). The Cl_2 gas has rapidly carried through the solution which is heated to the boiling point two-times. Next to the dilution with 15ml HCl, the solution is moved to a distillation flask that possess a dip tube, which forces the constant gas current to set off from the solution in the period of distillation.

During Cl_2 gas passage through the still, Ge has been distilled into a trap comprising 6N NaOH. Along with the HCl supplementations between evaporations, the solution has evaporated several times to reach the drying point. An overall volumetric range of 25-35ml has been distilled.

Subsequently, by adding 5ml of a saturated HCl solution of CuCl to the still and passing HCl gas through the still, As has distilled into an ice-cooled water trap. The distillation is resumed up till activity would not be distinguished in the distillate. The active distillate has dried to the wanted size, though Cl_2 gas bubbling into the solution.

X. Sr^{85}. TM: RbCl.

Prod.: 99 + %Sr^{85}, in HCl or HNO_3 solution.

Prod. Reac.: Rb^{85} (p,n) Sr^{85}; or Rb^{85} (d,2n) Sr^{85}.

OIR: Rb^{85} (d,α) Kr^{85}.

Chem. Sep. Princ.: Sr^{85} has carried on $Pb(NO_3)_2$ and in the time of sample dissolution Kr^{85} is volatilised. Then, Pb carrier is eliminated as PbS.

Rec.: 98%; TR: 3hrs.

The RbCl, about 5g, is dissolved in water, acidulated, and filtered out. 10mg Pb carrier are added and the solution condensed to 1ml by evaporation. 30mg of fuming HNO_3 are added and the solution cooled down in an ice bath, with infrequent blending.

$Pb(NO_3)_2$ which carries the Sr^{85} quantitatively, is eliminated by centrifugation, dissolved in a minimum volume of water, and reprecipitated. The $Pb(NO_3)_2$ is sorted out by centrifugation, and dissolved in water (25ml).

PbS is deposited by saturating the solution with H_2S. Sr^{85} is quantitatively reserved in the supernate. The solution is filtrated out, heated to an evaporation point near to dryness, and the activity solvated in HCl or HNO_3.

XI. Y^{88}. TM: strontium oxide (SrO).

Prod.: 98 + %Y^{88}, in HCl or HNO_3.

Prod. Reac.: Sr^{88} (p,n) Y^{88}.

OIR: Sr^{88} (p,2n) Y^{87}.

Chem. Sep. Princ.: Y^{88}, is carried on $Fe(OH)_3$. The Fe^{III} is eliminated by IEC.

Rec.: 98%; TR: ~4hrs.

The SrO target, about 5g, is dissolved in 0.1N HCl. 10mg of Fe^{III} carrier are added. CO_3^{-2} - free NH_4OH is added until the pH reaches 8.5. The $Fe(OH)_3$ precipitate, which carries Y^{88} quantitatively, is segregated by centrifugation. 4- reprecipitations are performed, using a minimum of HCl to dissolve the precipitates. The last precipitate is washed twice with water, then solvated in a minimum quantity of concentrated HCl (6N). The solution is moved across an Amberlite IRA-400 column that has been maintained with 6N HCl. The activity is washed from the column with 6N HCl. Then, the eluate is evaporated near to dryness then dealt with HNO_3 to break OM down. After that, the Y^{88} can be transformed to chloride by heating to dryness with HCl. It should be noted that if Al is present (from backing or container), several $Fe(OH)_3$ precipitations with high concentrations of NaOH should be performed.

XII. Cd^{109}. TM: Ag.

Prod.: 98 + %Cd^{109}, in HCl or HNO_3 solution.

Prod. Reac.: Ag^{109} (p,n) Cd^{109} or Ag^{109} (d,2n) Cd^{109}.

OIR: Ag^{107} (p,n) Cd^{107} or Ag^{107} (d,2n) Cd^{107}; Ag^{109} (d,α) Pd^{107}.

Citation: Aljerf L, Dastan D, Sajjadifar S, Bhatnagar S, Ukaogo PO, et al. (2019) Guidelines for the preparation and isolation of Radionuclides produced with In-house Cyclotrons Bombardments Open Journal of Chemistry 5(1): 020-029. DOI: http://dx.doi.org/10.17352/ojc.000013

Chem. Sep. Princ.: Cd^{109} is extracted from a thiocyanate (SCN^-) medium into chloroform ($CHCl_3$)-pyridine (C_5H_5N), back-extracted into 1M HNO_3, and this cycle repeated. Final purification is affected by means of anion exchange.

Rec.: 95%; TR: ~6hrs.

The active portion of the target, about 10g, is dissolved in HNO_3. After evaporating the solution to dryness, 150ml of water are added and any insoluble matter is eliminated by filtration. Solid NH_4CNS is then added until the precipitate which first forms are re-dissolved. The pH is adjusted to 5 with NaCNS, and the solution is transferred to a separatory funnel. The Cd^{109} is extracted 3-times with 50ml of $CHCl_3$ containing 50% of C_5H_5N. The activity is back-extracted into three 25ml portions of 1M HNO_3, which are then evaporated near to dryness. The activity is dissolved, treated, and extracted with $CHCl_3$- C_5H_5N, as before. After back-extracting into three 25ml portions of 1M HNO_3 and evaporation near to dryness, the activity is solvated in 0.3N HCl (10ml) and passed through a Dowex-1 column which has been washed with water and conditioned with 0.3N HCl. With six column volumes of 0.3N HCl, the column is rinsed, after which the Cd^{109} is eluted with 0.3mN HCl. The active eluate is evaporated near to dryness and dealt to break OM down. By evaporation with HCl, Cd^{109} may be transformed to chloride.

XIII. In^{111}. TM: Cd.

Prod.: 98 + % In activities, in HCl or HNO_3 solution.

Prod. Reac.: Cd^{110} (d,n) In^{111}; Cd^{111} (d,2n) In^{111}.

OIR: Cd^{108} (d,n) In^{109}; Cd^{110} (d,2n) In^{110}; Cd^{113} (d,n) In^{114}; Cd^{114} (d,n) In^{115}; Cd^{116} (d,2n) In^{116}; Cd^{116} (d,n) In^{117}.

Chem. Sep. Princ.: In-activities are carried on $Fe(OH)_3$ from ammoniacal solution. The Fe is extracted with isopropyl ether ($(CH_3)_2CHOCH(CH_3)_2$).

Rec.: 98%; TR: ~4hrs.

The Cd target metal, about 10g is etched from its backing with HCl. 20mg of Fe^{III} carrier are inserted, and the solution is neutralised with NH_4OH. 20ml of excess NH_4OH are added. The deposition layer is segregated by centrifugation and solvated in a min. quantity of HCl. After diluting to 15ml with water, the precipitation is repeated. After two additional reprecipitations, 15ml of 8N HCl are added, and the Fe extracted 3-times with $(CH_3)_2CHOCH(CH_3)_2$. The liquid is evaporated near to dryness. With HNO3, OM is broken down by repeated evaporations. By evaporation with HCl, the transformation to chloride can be affected.

XIV. Ce^{139}. TM: lanthanum oxide (La_2O_3).

Prod.: 98 + %Ce^{139}, in HCl or HNO_3 solution.

Prod. Reac.: La^{138} (d,n) Ce^{139}; La^{139} (d,2n) Ce^{139} or La^{139} (p,n) Ce^{139}.

OIR: none.

Chem. Sep. Princ.: Ce^{139} is extracted into hexone (4-methyl 2-pentanone).

Rec.: 95%; TR: ~2hrs.

The La_2O_3 target, about 3g, has been dissolved in a min. quantity of 3N HCI. 5ml of HNO_3 are added and the solution is dried by evaporation. A blend of conc. HNO_3 (6ml), 2M $NaBrO_3$ (2ml), and water (2ml) are added, and the resultant solution is thoughtfully heated until a vigorous reaction ensues. The solution is cooled out and moved to a separator funnel comprising of 50ml of hexone which has been conditioned by agitation with 50ml of 9M HNO_3. 1ml of 2M $NaBrO_3$ and 1.5 ml of HNO_3 are inserted. The funnel is instantly agitated, the phases permitted to discrete, and the aqueous layer retained for next retrieval of Ce^{139}. It is highly vital to have bonus amount of $NaBrO_3$ existing during extraction. The hexone layer is rinsed two-times with 9M HNO_3 (10ml) and a few drops of 2M $NaBrO_3$. The washes are reserved. Ce^{139} is then extracted from the hexone with double quantity of 5ml portions of water comprising 4 drops of H_2O_2 (30%). The hexone is castoff into a stream of flowing water. Attention: HNO_3 – hexone is possibly explosive.

The remaining Ce^{139} in the aqueous phase formerly retained has extracted with hexone, and subsequently the aqueous phase is eliminated. The hexone layer is washed with double quantity with 9N HNO_3 and Ce^{139} is eliminated from the hexone with double portions of water (5ml) encompassing 30% H_2O_2 (4 drops). The hexone is removed. Since the scattering of Ce^{139} amongst the hexone and 9N HNO_3 washes is nearly to 8:1, it is required to retrieve the Ce^{139} in the 4-rinses. These are aggregated and evaporated till dryness. A blend of conc. HNO_3 (6ml), 2M $NaBrO_3$ (2ml), and water (2ml) is added, and the ending solution heated. The Ce^{139} in the solution is extracted twice with hexone. The hexone layers are each washed two-times with 9M HNO_3 (10ml), and the washes are discarded. The Ce^{139} in the hexone phases is then back-extracted with double 5ml portions of water comprising 30% H_2O_2 (4drops). The later oxide extracts are combined with the first H_2O_2 extracts and are then evaporated near to dryness. OM has broken down with HNO_3. The transformation to chloride can be fulfilled by evaporation with HCl.

XV. W^{181}. TM: tantalum (Ta).

Prod.: 98 + %W^{181}, in HCl-HF.

Prod. Reac.: Ta^{181} (p,n) W^{181} or Ta^{181} (d,2n) W^{181}.

OIR: none.

Chem. Sep. Princ.: W^{181} is isolated apartly from Ta by IEC. The last separation is completed by coprecipitating W^{181} with $Fe(OH)_3$ at pH 4. Fe is extracted with $(C_2H_5)_2O$.

Rec.: 90 +%. TR: ~10hrs.

In a platinum (Pt) dish, the Ta target is etched with HF to eliminate the active layer. Approximate 5g are solvated. After the evaporation near to dryness, HF (4ml) and HCl (20ml) are added, and the blend is heated to get a pure solution. A volume of water (16ml) is added. The solution is passed across a 40cm polyethylene (PE) column containing Dowex-1 which has been conditioned with 1.5N HF + 6N HCl. W^{181} is eluted with 1.5N HF + 6N HCl. Fractions of 20ml are brought in PE containers. The active fractions are integrated and dried by evaporation to 5ml in a Pt dish. Water (30ml), boric acid (H_3BO_3) (200mg), and Fe^{III} carrier (10mg) have been supplemented. Adequate 6N NaOH is inserted to reach pH 4.0. The resultant precipitate is segregated by centrifugation and solvated in a min. quantity of conc. HCl. The precipitation is reiterated more than two-times. The ending precipitate is rinsed out two-times with water and solvated in 6N HCl. With $(C_2H_5)_2O$, Fe has extracted 3-times. The aqueous phase is heated and evaporated near to dryness. By evaporation with HNO_3, the OM is broken down. Whereas, by evaporating with HCl, the transformation to chloride is affected. One droplet of HF is added.

XVI. Bi^{207}. TM: Pb.

Prod.: 98 + %Bi activities, in HNO_3 solution.

Prod. Reac.: Pb^{207} (p,n) Bi^{207}; Pb^{208} (p,2n) Bi^{207}.

OIR: Pb^{206} (p,n) Bi^{206}; Pb^{207} (p,2n) Bi^{206}.

Chem. Sep. Princ.: The Pb is eliminated by precipitation with fuming HNO_3. Moreover, in the incidence of an extra amount of NaOH, Bi is coprecipitated with $Fe(OH)_3$. After that, Fe is taken off through the extraction with $(C_2H_5)_2O$.

Rec.: ~90%; TR: ~6hrs.

Approximately 25g of the Pb target metal, is etched from its backing with 6N HNO_3. The ending slurry is moved to a centrifuge bottle after evaporating to around 50ml, then fuming HNO_3 (100ml) is added. Immediately, the precipitate is eliminated by centrifuge technique after cooling in an ice bath and then rinsed two-times with cold fuming HNO_3. Fe^{III} carrier (20mg) is added to the aggregate mixture of the supernate and washes, and the solution is evaporated near to dryness. A volume of water (15ml) is added; then 9N NaOH (15ml) is supplemented and the blend agitated. Bi is quantitatively transferred by the $Fe(OH)_3$, whereas the solvable plumbate (formed by the reaction of lead(*IV*) oxide, PbO, with alkali) stays in the supernate. The sedimentation is separated by centrifugation and solvated in a min. quantity of conc. HCl. 3-reprecipitations are carried out. The ending precipitate is solvated in 6N HCl (15ml), and with $(C_2H_5)_2O$, the Fe is extracted 3-times. OM is eliminated by evaporation with HNO_3.

Conclusion

The current study is significant for the producers, processers and distributers of carrier-free radioisotopes and cyclotron-produced. Even though, cyclotron produced radioisotopes are intrinsically considered of extremely higher-priced compared to the reactor-produced radioisotopes, numerous conditions have been suggested under which their applications are preferable, and for such circumstances sound obligatory. Quite a lot of new isotopes, formerly rendered hard to get, are recently effortlessly obtainable. Our suggested measures have been described in more details for the partition of seventeen radioisotopes in a carrier-free shape: Be^7, Na^{22}, V^{48}, Mn^{52}, Mn^{54}, Fe^{55}, Co^{57}, Zn^{65}, Ga^{67}, As^{74}, Sr^{85}, Y^{88}, Cd^{109}, In^{111}, Ce^{139}, W^{181} and Bi^{207}. In addition, yield-dense targets have been designed for the synthesis of these radioisotopes by means of the cyclotron irradiations by using 15MeV energy of deuterons in association with other protons' energies (8, 15 and 20MeV).

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