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Radiochemical Separation Relevant to the no-carrier-added Production of ⁹⁰Nb: A Potential Radiopharmaceutical for PET Imaging

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ABSTRACT

Niobium-90 (⁹⁰Nb), a radioisotope of paramount importance in the field of nuclear medicine, has been effectively synthesized and separated from target materials through natZr(p,n)90Nb and 90Zr(p,n)90Nb reactions. The attainment of high-purity Niobium -90 necessitates the use of exceptionally pure zirconium-90 isotopes, prompting the establishment of a meticulously structured three-stage production process. In the initial phase, the enrichment of ⁹⁰Zr stable is achieved through Electromagnetic Isotope Separation (EMIS). The resulting enriched zirconium oxide target material undergoes rigorous validation through X-ray Diffraction (XRD) analysis, confirming isotopic and chemical purities quantified at 99.22% and 99.85%, respectively. These purities are ascertained through advanced techniques, including gamma spectrometry and Particle-Induced X-ray Emission (PIXE). The subsequent stage involves the irradiation of target materials, prepared from natZrO₂ and 90ZrO₂ powders, within the cyclotron accelerator. The third and final phase, post-irradiation, encompasses an elaborate chemical purification process, employing ion-exchange method. This process refines Niobium -90 from the target materials. The assessment of Niobium-90 activity purity, derived from both natural and enriched sources, confirms purities of 98.69% and 100%, respectively, through meticulous examination using a High Purity Germanium (HPGe) detector.

GRAPHICAL ABSTRACT



Introduction

(⁹⁰Nb) Niobium-90 is а radioisotope distinguished by its remarkably brief half-life of 6.14 hours and a substantial positron emission rate of 53%, rendering it a pivotal asset in the domain of positron emission tomography (PET) imaging. Its multifaceted applications in the field of nuclear medicine encompass vital roles, including the detection and assessment of tumor activity, as well as the investigation of fundamental physiological processes within living organisms [1-2]. With its low-energy beta radiation (E_{mean}=350 keV), ⁹⁰Nb is well-suited for immuno-PET, facilitating the examination of antibodies and their fragments. Its capability for high-resolution imaging enhances its utility in the realm of biological and medical research [3-**4**].

The predominant techniques employed for the production of ⁹⁰Nb predominantly revolve around nuclear reactions, notably ^{nat}Zr(p,xn)⁹⁰Nb and ⁹⁰Zr(p,n)⁹⁰Nb, conventionally executed through the irradiation of Zirconium metal with protons using a cyclotron accelerator [5]. Historically, researchers have favored the utilization of slender foils crafted from natural Zirconium metal as the principal targets for these nuclear reactions [6]. Nevertheless, a relatively uncharted avenue within this domain pertains to the exploration of metallic targets derived from

enriched Zirconium. Presently, the prevailing methodology primarily centers on the adoption of compressed disc-shaped targets synthesized from Zirconium oxide powder [7].

The primary objective of isotope enrichment within a chemical reaction is to enhance the yield of the targeted radiopharmaceutical while simultaneously mitigating the inadvertent generation of undesirable radio nuclides [8-9]. This necessitates the utilization of diverse techniques for isotope separation, including gas centrifugation, laser isotope separation, Ion Cyclotron Resonance (ICR) in conjunction with cyclotron-produced plasma, plasma centrifugation, and electromagnetic separation. Of these methods, ICR has emerged as an industrially applicable approach for the extensive production of stable isotopes [10]. The electromagnetic isotope separator an is apparatus utilized for the separation of stable isotopes of various elements using a magnetic field to differentially segregate ions with distinct mass but consistent energy levels. This technique has the potential to yield isotopes with exceptionally high levels of enrichment [11-12]. Subsequent to the isotope separation process, the ensuing stage in target enrichment involves the elimination of impurities from the intended isotope through the employment of chemical methodologies like electrolysis and ion-exchange resins, among others. Conventionally, the desired target is synthesized in the form of isotope oxides, with the chemical purity thereof ascertained through techniques such as Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES), while the isotopic enrichment is determined utilizing Mass Spectrometry (MS).

Following the target enrichment process, the enriched target is subjected to precisely formulated reactions with the explicit goal of producing the intended product. Significantly, the isotopic and chemical purity of the resulting radioisotope holds paramount importance. Indeed, as the purity of desired product (in this case, 90Nb) increases and impurities decrease, the level of radiation absorption in healthy tissues decreases. Thus, it is imperative to engage in chemical purification processes and employ separation techniques for discerning the reaction product from the target. These techniques encompass an array of methods, including solvent extraction [13], silica gel [14-16], and anion-exchange resins [17].

Within this study, the production of high-purity Niobium-90 entailed the following sequential stages:

Commencing with the preparation of a highpurity Zirconium-90 target isotope, it is achieved through electromagnetic separation. Subsequently, the removal of impurities was executed via an electrolysis process, employing alpha-hydroxy phenyl acetic acid for the precipitation of zirconium ions. Ultimately, highpurity Zirconium-90 oxide powder was attained, serving as the intended target for the ensuing reaction. The isotopic and chemical purity of this prepared target was meticulously validated using Particle-Induced X-ray Emission (PIXE) and mass spectrometry.

The prepared targets, derived from ^{nat}ZrO₂ and ⁹⁰ZrO₂ powders, were enclosed within aluminum foils and introduced into a cyclotron accelerator (Cyclone-30, IBA Belgium), subjected to proton bombardment to yield Niobium -90. Subsequent to the reaction procedures, Niobium-90, the radioisotope, was effectively isolated from the

natural and enriched targets, with the aid of anion-exchange resins, facilitating the subsequent evaluation and conclusive verification of its isotopic purity and activity percentage via an HPGe detector.

Each of the aforementioned procedural steps will be expounded upon in a comprehensive manner in the forthcoming sections of this scholarly investigation.

Experimental

Materials

The initial material chosen for this process is a chemical compound of the target element, such as Zirconium tetrachloride. Pure Zirconium is not practical due to its high melting point. For enrichment process, we procured pure copper sheets, high-purity graphite plates, Zirconium tetrachloride (ZrCl₄) with a purity level exceeding 98%, mandelic acid, nitric acid, hydrochloric acid, all sourced from Merck Company, Germany.

Devices instrument

In the course of conducting experiments and gathering essential data to ensure the successful outcomes of our research project, we employed a range of specialized equipment and instrumentation. These included:

Electromagnetic Isotope Separator System (CI-140 Model, China): This system played a pivotal role in our research, facilitating the isotopes separation using electromagnetic fields.

X-ray Diffraction Instrument (W-Ka X-ray Source Stol Stidy-Mp Diffractometer Phillips Expert Model, Netherland): We utilized this precision instrument to analyze the crystallographic properties of our materials and gain insights into their atomic structure.

Electrostatic Accelerator-Based Proton-Induced X-ray Emission (PIXE) System (Van de Graaf accelerator of high voltage engineering, USA) with 2 MeV Proton Beam: This system allowed us to perform elemental analysis with exceptional precision, providing valuable information about the composition of our samples. Mass Spectrometer (LZD-207 Model, China): Mass spectrometry was a crucial tool in our research, enabling us to determine the precise mass-to-charge ratios of ions and molecules in our samples.

Furnace (Kaviran Industrial Furnace, KIF 1200, Iran): The furnace was instrumental in creating controlled environments for high-temperature experiments, ensuring the accuracy of our findings. The effective utilization of these advanced instruments and systems greatly contributed to the success of our research endeavors.

Electromagnetic isotope separation of Zirconium isotopes

Zirconium possesses five stable isotopes: ⁹⁶Zr, ⁹⁴Zr, ⁹²Zr, ⁹¹Zr, and ⁹⁰Zr, with natural abundances of 2.76%, 17.28%, 17.19%, 11.32%, and 51.45%, respectively. In this work, we enriched the ⁹⁰Zr isotope using the electromagnetic isotope separation (EMIS) method. In this approach, the commences process with the thermal evaporation of atoms from the target element within an ion source known as a calutron. Subsequently, these atoms undergo ionization via electron collisions within the ionization chamber. Once ionized and subjected to a uniform magnetic field with constant energy, they traverse a semicircular trajectory. Owing to the unchanging energy of ions and the consistent magnetic field strength, the trajectory radius varies according to their respective masses. Consequently, upon the culmination of a single operational cycle, isotopes are extracted from the packets situated on the collector [18]. Figure 1 depicts the frontal view of the graphite plate in the electromagnetic separation system, along with the copper pockets positioned on the collector, prior to the commencement of the separation process.

Throughout the separation process, both physical and electrical parameters should be methodically calculated and determined, both prior to and during the separation of each element. Subsequent to the preparation of initial materials, the redesign of the collector, and the fabrication of copper packets, a quantity of 50 grams of Zirconium tetrachloride (ZrCl₄) raw material was loaded into the graphite crucible within the ion source. Table 1 presents the operational parameters defined for the electromagnetic isotope separator system applied in the separation of zirconium isotopes. Following the fine-tuning of relevant parameters and attainment of the desired conditions, the process yielded the Zirconium isolated isotopes, which were collected in the collector.





Figure 1: (a) The front panel of graphite and (b) copper pockets installed on the collector

VAccelerator.	VFocus.	IFilament	VCathode	ICathode	VAnode	IAnode	IMagnet	Vacuum	Temperature	I _{Total}
(keV)	(keV)	(A)	(V)	(A)	(V)	(A)	(A)	(Pa)	(°C)	(mA)
30.8	10.2	63	700	0.48	163	0.83	123	3 x 1 0 - 3	135	11

Table 1: Operational parameters during the separation of Zirconium isotopes

Chemical purification of ⁹⁰Zr isotope

Given that Zirconium ions are collected on the surface of the copper packet, the primary impurities associated with Zirconium are copper ions. Typically, the extraction of enriched isotopes from copper packets involves the use of nitric acid. As a result, the initial step involves the extraction of the ⁹⁰Zr isotope, which has been collected on the copper packet, using a 50% nitric acid solution. Following electrolysis and subsequent detailed procedures discussed in later sections, enriched zirconium oxide was obtained, serving as the irradiation target to produce ⁹⁰Nb.

Faced challenge and potential sources of error

Given the constrained availability of enriched isotopes, it is crucial to prevent any depletion of ⁹⁰Zr. To confront this challenge, the initial implementation of chemical separation processes was directed at isotope 92Zr. Subsequently, these procedures were adapted and improved for the separation of 90Zr, informed by the experiences gained from working with ⁹²Zr. In other aspects, no specific challenges were encountered, provided that meticulous care was exercised in handling chemical substances. The primary source of error in the separation process is the presence of impurities, including isotopic and non-isotopic impurities. Through meticulous attention to detail and process optimization, these errors can be mitigated to a satisfactory extent.

Potential radiation risks

In order to mitigate the risk of radiation exposure during the irradiation and separation of the radioactive nuclide ⁹⁰Nb, the following precautions were taken. All pertinent equipment was situated within hot cells located in the cyclotron facility. Containers made of teflon and non-glass materials were exclusively chosen, and lead shielding was employed during their transportation and manipulation. The separation process was executed in a semi-automated manner within shielded hoods and lead-lined walls. By implementing these measures and finetuning separation parameters, the potential for radiation hazards was significantly reduced.

Chemical purification and extraction of Niobium from Zirconium

Following irradiation, the extraction and separation of Niobium-90 produced from Zirconium targets were accomplished using an ion exchange method employing an anion exchange resin (Cl⁻ (200-400 mesh, Dowex 1-X8). An anion exchange resin such as RN+(CH₃)₃Cl-, where R represents the resin base, possesses non-mobile cationic sites to which mobile anions Cl- or OH- can be attached. Through ion like exchange, the cations or anions present in the solution are exchanged with the cations and anions present on the resin. This exchange occurs in a way that both the solution and the resin remain electrically neutral. This is achieved through solid-liquid equilibrium without the solid dissolving in the solution. Ion exchange is an equilibrium phenomenon that can generally be described for a cation exchange system by the following equation:

$$R_{n}^{-}M^{n+} + nH_{s}^{+} \rightarrow n(R^{-}H^{+})_{r} + M_{s}^{n+}$$
 (1)

The degree of ion adsorption by a resin is quantified as the distribution coefficient, symbolized as K_d , and its calculation is based on the following equation:

$$K_d = \frac{(C_A)_r}{(C_A)_s} \tag{2}$$

In which $(C_A)_r$ and $(C_A)_s$ are the concentrations of ion A in the resin and in the solution respectively. The greater the distribution coefficient (K_d) of an ion, the more robust its affinity for the resin. Consequently, by assessing the distribution coefficients (K_d) of elements associated with a particular resin, it becomes feasible to effectively segregate these elements from one another. When the disparities in K_d values among elements are substantial, the separation process is notably enhanced [19]. Given that chloride ions serve as the active agents of the resin, a procedure known as "resin conditioning" was executed. In this process, the resin was submerged in a stagnant 28 M hydrofluoric acid solution for 24 hours. This immersion aimed to displace the chloride ions with fluoride ions. Following this conditioning process, the prepared resin was employed for the subsequent separation process.

Results and Discussion

Separation of Zirconium isotopes

Figure 2 visually depicts the resolved spectrum of Zirconium isotopes, meticulously recorded by a Faraday cup positioned on the collector's door. As evident, the peaks exhibit clear separation, with their heights directly proportional to the isotopic abundances. With the precision of the separation method verified, the isotopes were systematically collected into packets.

Chemical purification of ⁹⁰Zr isotope

Figure 3 depicts an illustration of the washing process for the ⁹⁰Zr isotope packet utilizing nitric acid. To eliminate copper ions effectively, electrolysis was employed under a constant current of 2 Amperes. This process resulted in the deposition of metallic copper on the surface of a platinum electrode utilized as the cathode. The electrolysis continued until the blue color associated with copper ions disappeared. Figure 4 visually presents various stages of electrolysis and the resulting color transformation of the solution.



Figure 2: The spectrum recorded by the detector during the sweeping of accelerator voltage



Figure 3: Washing the ⁹⁰Zr isotope packet



Figure 4: The color of the final solution (a) before electrolysis and (b) after electrolysis

Precipitation of Zirconium Ions and Impurities as Hydroxides

Following the solution electrolysis, all Zirconium ions, along with other undesired impurities, were selectively precipitated as hydroxides. This was accomplished by the addition of concentrated ammonia solution.

Dissolution of Hydroxide Precipitate

In the subsequent phase, the resulting hydroxide precipitate was dissolved in a solution consisting of 20 mL of 6 molar hydrochloric acid. To achieve the desired concentration, 20 mL of distilled water was added, resulting in a final concentration of 3 molar.

Heating and Agitation

The resulting Zirconium chloride solution was heated to 85 °C on a hot plate, with continuous agitation, for 20 minutes.

Concurrently, 250 mL of a 16% weight/volume solution of mandelic acid (prepared by dissolving 80 grams of mandelic acid in 500 mL of distilled water) was added into the solution.

Extended Heating

The resulting mixture was then heated at 85 °C on a hot plate for an additional 40 minutes. Under these conditions, a milky precipitate of Zirconium mandelate was formed.

Settling and Separation

The Zirconium mandelate precipitate was allowed to settle near the mother solution overnight. Subsequently, it was effectively separated from the solution using a centrifuge.

Drying

Finally, the Zirconium mandelate precipitate was subjected to drying at 110 °C, as illustrated in Figure 5.

Utilization of Mandelic Acid Solution



Figure 5: (a) The dissolution of Zirconium hydroxide in hydrochloric acid, (b) formation of milky white color Zirconium mandelate, and (c) separated Zirconium mandelate after drying at 110 °C

Table 2. 110perfies of Zircolnulli and Zircolnulli oxide						
Specifications	Zirconium	Zirconium oxide				
Atomic weight (g/mol)	91.224	123.22				
Density (g/cm ³)	6.52	5.68				
Melting point (°C)	1855	2715				
Thermal conductivity (300 K) (W/(m⋅K))	22.6	2.2				

Table 2: Properties of Zirconium and Zirconium oxide



Figure 6: (a) The enriched Zirconium oxide powder serves as the final product and (b) the weight of the obtained product

To ensure the final product maintains a consistent composition and weight, it's necessary to convert Zirconium mandelate into Zirconium oxide (⁹⁰ZrO₂) through a heat treatment process in a furnace. The thermal energy generated during the interaction of charged particles with the target material can elevate its temperature significantly. Under these conditions, there is a risk of target material damage or chemical alterations. Utilizing target materials in their solid phase is more prevalent due to increased activity production, improved heat transfer characteristics, and the ability to control potential contamination leakage.

Zirconium oxide, as one of the compounds of Zirconium, possessing a high melting point and remarkable stability, can serve as a suitable target material. A summary of the properties of Zirconium metal and Zirconium oxide is provided in Table 2.

The Zirconium mandelate underwent a crucial heat treatment process within a furnace, where it was exposed to a temperature of 1000 °C for 6 hours. As a result of this controlled treatment, a total of 690 milligrams of enriched Zirconium oxide powder was successfully produced, as illustrated in Figure 6.

Figure 7 presents the X-ray diffraction (XRD) spectrum associated with the analysis of ⁹⁰ZrO₂. In this figure, the scattering peaks observed at (011), (-111), (111), (002), (200), (-211), (022), (-220), (-113), and (131) are indicative of the formation of Zirconium oxide with a monoclinic structure, consistent with reference number 0420-036-00. This confirms the production of Zirconium-90 oxide.



Figure 7: The XRD spectrum associated with the analysis of ⁹⁰ZrO₂

Table 3 presents data regarding the isotopic purity of Zirconium-90, represented as the abundance percentage of the enriched isotope, achieved through the separation process utilizing mass spectrometry. This data is included for comparison with empirical results obtained from earlier experiments. In this investigation, the isotopic enrichment of Zirconium-90 has been quantified with a relative error of 0.03.

The chemical purity of ⁹⁰Zr produced in this particular study has been comprehensively evaluated, and the results are presented in Table 4. According to the findings, the chemical purity of the ⁹⁰Zr was exceptionally high, as determined to be 99.85% through Proton-Induced X-ray Emission (PIXE) analysis.

Proton bombardment of natural and enriched Zirconium targets

Targets in the form of discs were meticulously prepared using powders of ⁹⁰ZrO₂ and natZrO₂. These targets were designed for irradiation with proton beams, and their production involved the use of a press apparatus. Subsequently, for the purpose of producing Niobium-90, the prepared samples were subjected to irradiation. This irradiation process took place at the Alborz Health and Agriculture Research Center and utilized proton beams with energies of 17.7 and

Abundance percent of enriched	Method	Ref.	
00.22		Oran ang sh	
99.22	Electromagnetic separation	Our work	
99.2	Electromagnetic separation	[23]	
97-99	Electromagnetic separation	[22]	
98.00	Electromagnetic separation	[24]	
90-98	Gas centrifugation	[10]	

Table 3: Comparing the isotopic purity of Zirconium-90 achieved in the present study with empirical findings

Table 4: The results related to the chemical purity analysis of enriched Zirconium using PIXE method

Chemical compounds	⁹⁰ ZrO ₂	ZnO	Cu ₂ O	Fe ₂ O ₃	CaO
Percentage	99.85	-	-	0.05	0.10

21.5 MeV. These beam energies had been determined as optimal for Niobium-90 production based on theoretical calculations [5]. The irradiation procedure was conducted with a beam current of 5 μ A and lasted for 60 minutes. Following the irradiation phase, the target discs were allowed to cool down in the hot cell of the cyclotron facility. Here, they were allowed to cool for a period of 24 hours. This cooling period was essential for allowing the short-lived activities of the target disc shell to naturally decay. After the cooling period had elapsed, the target discs were removed from the shuttle and meticulously prepared for the subsequent stages of chemical purification and Niobium extraction from the Zirconium target.

The *nat*Zr(p, xn) reaction

The process for the dissolution and separation of the target materials from bombarded natural Zirconium oxide was executed meticulously, as outlined below:

Sample Preparation: Initially, 180 milligrams of bombarded natural Zirconium oxide were placed in a Teflon beaker.

Addition of Hydrofluoric Acid: To initiate the dissolution process, 5 mL of 28 molar hydrofluoric acid were added to the beaker.

Heating and Stirring: The mixture was placed on a heater and stirred using a magnetic stirrer. It was heated to 120 °C and maintained for 15 minutes to ensure complete dissolution.

Column Separation (Sample No. 1): The resulting solution was then passed through a column containing 4 grams of anion exchange resin (Dowex 1-X8, Cl-) using a peristaltic pump. This step took approximately 10 minutes, and the output was designated as Sample No. 1, which was stored in a Teflon container.

Elution of Impurities (Sample No. 2): To remove any residual Zirconium and other impurities like Yttrium from the column and connectors, a volume of 20 mL of 28 molar hydrofluoric acid was passed through the column for 15 minutes. The output of this process was labeled as sample No. 2 and stored separately in another container. *Niobium Extraction (Sample No. 3)*: During this stage, it was anticipated that all Zirconium and other impurities would have been removed from the column, leaving only Niobium remaining. To extract Niobium from the column, a mixture of 6 molar hydrochloric acid and 2.44% oxygenated water was utilized. Therefore, 20 mL of the specified solvent mixture was passed through the column for 15 minutes, and the output was labeled as sample No. 3, treated as a separate entity.

The entire dissolution and separation process, from the initial dissolution of the sample to the extraction of Niobium, was carried out efficiently within a total duration of 55 minutes.

The rationale behind passing 28 molar hydrofluoric acid through the column stems from the findings of studies on the adsorption behavior of different ions within a concentration range of 1 to 24 M hydrofluoric acid, as exemplified by Faris curves [25]. These studies have revealed distinct trends in the adsorption behavior of Zirconium and Niobium as a function of hydrofluoric acid concentration. Specifically, the research has indicated that as the hydrofluoric acid concentration increases, the Zirconium adsorption decreases. Conversely, the Niobium adsorption reaches its maximum at a concentration of 24 molar hydrofluoric acid. Moreover, complementary studies conducted by Kraus-Moore have investigated the adsorption behavior of various ions within a concentration range of 1 to 12 molar hydrochloric acid [26]. These investigations have unveiled that the most efficient extraction of Niobium is achieved using 6 molar hydrochloric acid. It is of paramount importance to underscore that the optimization of solvent quantities, temperature, and duration will play a substantial role in mitigating error levels.

In the subsequent step, radio isotopic purity was assessed using High Purity Germanium (HPGe). For this purpose, specific volumes of various solutions were subjected to analysis. The volumes of the solutions analyzed were as follows: Solution before Separation: 20 microliters, Solution Number 1: 60 microliters,



Figure 8: The gamma spectrum obtained from the sample prior to separation and the characteristic peaks

Solution Number 2: 60 microliters and Solution Number 3: 80 microliters. The results of these analyses have been visually represented in Figures 8 to 11, presented in the respective order. As observed, when utilizing proton energy of 21.5 MeV, the irradiation process not only yields Niobium-90 but also results in the production of radioisotopes like Zirconium-89 and Yttrium-87. However, a meticulous spectroscopic analysis of the sample derived from target dissolution does not reveal the existence of radioisotopes stemming from the target shield material. Consequently, the presence of radioactive Zirconium-89 within the target material can be employed as a reliable indicator, affirming the successful chemical separation of Niobium from the Zirconium target.

The gamma spectrum of passing solution number 1, illustrated in Figure 9, provides important information. It is clearly evident from the spectrum that all of the produced Niobium was effectively retained on the column, as there are no discernible Niobium peaks present in the spectrum of the output solution. This outcome aligns with the desired goal of retaining Niobium for subsequent extraction while allowing other elements to pass through. Similarly, the spectrum of the output solution number 2, as demonstrated in Figure 10, also demonstrates the absence of Niobium peaks. This result is consistent with the successful separation and retention of Niobium. However, both spectra reveal slight peaks corresponding to radioactive Zirconium-89 and Yttrium-87. These peaks are a direct result of the bombardment process and the subsequent production of these isotopes.

The separation of a small amount of active Zirconium-89, used as an indicator, serves as a clear confirmation of the successful elimination of Zirconium from the final Niobium product. The gamma spectrum of the Niobium product generated from natural Zirconium, represented by sample number 3, is presented in Figure 11. In this spectrum, it is evident that Zirconium-89 and Yttrium-87 have been effectively removed, leaving only Niobium isotopes present.



Figure 9: The gamma spectrum obtained from sample number 1 after passing through the column



Figure 10: The gamma spectrum obtained from sample number 2 after washing the column with HF to completely remove Zirconium and other impurities



Figure 11: The gamma spectrum of sample number 3, containing ⁹⁰Nb derived from natural Zirconium along with active impurities without chemical impurities. As seen, only the isotopes of Niobium remain.

The existence of a prominent gamma peak attributed to the radioisotope ⁹²Nb is quite evident in comparison to other radioisotopes. This specific radioisotope originates from the isotope ⁹²Zr through the reaction ⁹²Zr(p,n)⁹²Nb. Notably, because there is a substantial energy overlap range with the production of ⁹⁰Nb, the sole effective method to prevent its generation is by employing enriched ⁹⁰Zr as the target material.

The ⁹⁰Zr(p, n) reaction

In the subsequent step, 690 milligrams of enriched Zirconium oxide (90 ZrO₂), which had undergone bombardment in the previous stage, were placed into a Teflon beaker, following a procedure similar to that used for the natural sample. Subsequently, 5 milliliters of 28 molar hydrofluoric acid were added to the beaker. The mixture was stirred using a magnetic stirrer on a heater and heated to 120 °C for 15 minutes to ensure complete dissolution. Following the dissolution process, the sample was subjected to analysis to determine its radio isotopic purity, utilizing HPGe. The resulting gamma spectrum, specifically related to the radioisotope ⁹⁰Nb produced from enriched Zirconium with an enrichment level of 99.22%, is presented in Figure 12.

As evident, the radioisotopes ⁹²Nb, ⁸⁹Nb, and ⁹⁵Nb, which were detected during production with natural Zirconium, have been successfully eliminated. Consequently, the desired product is now free from radioactive impurities. The results of the purity percentages for ⁹⁰Nb activity produced in both reactions are presented in Table 5 and have been compared with the available experimental results.



Figure 12: Gamma spectrum of Niobium obtained from enriched zirconium without any radioactive impurity **Table 5:** Comparison of the ⁹⁰Nb activity purities for production by natural and enriched Zirconium targets

Reaction	Activity purity percentage of Niobium before chemical senaration	Activity purity percentage of Niobium after chemical senaration	Activity purity percentage of ⁹⁰ Nb after chemical senaration	Ref.
^{nat} Zr(p, xn) ⁹⁰ Nb	99.97	100	98.69	Our work
⁹⁰ Zr (p, n) ⁹⁰ Nb	-	-	100	Our work
^{nat} Zr(p, xn) ⁹⁰ Nb	-	99	-	[17]
^{nat} Zr(p, xn) ⁹⁰ Nb	-	99.89	97	[6]

As evident, the results of this study are highly remarkable. As a significant outcome, the elevated purity level of ⁹⁰Nb plays a pivotal role in enhancing its clinical utility. This heightened purity leads to enhanced image quality and a reduction in radiation exposure risk to healthy tissues, thereby optimizing its clinical applications.

Conclusion

The chemical separation processes resulted in highly purified ⁹⁰Zr stable. The chemical purity of ⁹⁰Zr was determined to be 99.85%, with an

isotopic purity of 99.22%, showing remarkable results when compared to past efforts.

An ion-exchange method effectively removed radioactive impurities, leading to the production of ⁹⁰Nb with 100% radioisotopic and activity purities, free from any radioactive impurities. These results hold great significance as they demonstrate the viability of producing ⁹⁰Nb using this method. The high purity levels achieved are promising and pave the way for potential replication and large-scale manufacturing. The successful production of ⁹⁰Nb opens the door to potential applications in clinical and medical fields, particularly in the context of PET imaging systems. Further research can focus on optimizing the production process, improving yields, and exploring additional applications for ⁹⁰Nb in the biological and medical domains. In addition, investigating the feasibility of largescale production will be a crucial next step.

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