Comprehensive theoretical studies on stable and radioactive isotopes produced by proton irradiation of titanium target

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Abstract. Natural titanium (\textsuperscript{nat}Ti) has been suggested to be used as a window or target body in cyclotron-based medical radionuclide production. Therefore, comprehensive theoretical studies regarding impurities that might contribute to proton bombardment of Ti window are of great interest. In this investigation, the TALYS code was employed to calculate nuclear cross-sections of \textsuperscript{nat}Ti(p,n), \textsuperscript{nat}Ti(p,\alpha), \textsuperscript{nat}Ti(p,\alpha), \textsuperscript{nat}Ti(p,d) and \textsuperscript{nat}Ti(n,\gamma) reactions. The calculations were simulated for proton energies between 1 to 60 MeV. Based on the calculated results, several vanadium (V) radionuclides, e.g. \textsuperscript{46,47,48,49,50}V were predicted to be generated from \textsuperscript{nat}Ti(p,n) reaction, whereas vanadium (V) radionuclides, namely \textsuperscript{45}V could be produced from \textsuperscript{nat}Ti(p,\alpha) reaction. Some scandium radionuclides, e.g. \textsuperscript{43,44,46,47Sc} could be resulted from \textsuperscript{nat}Ti(p,\alpha) reaction, while stable \textsuperscript{45}Sc isotope was also predicted from this reaction. For \textsuperscript{nat}Ti(p,d) reaction, \textsuperscript{45}Ti radionuclide was expected to be generated, while other stable \textsuperscript{46,47,48,49}Ti atoms could be created as a result of this nuclear reaction. In addition, stable isotope \textsuperscript{46}Sc was expected to be produced from \textsuperscript{46}Ti(p,2p)\textsuperscript{45}Sc reaction, while \textsuperscript{47,48,49}Sc radionuclides were foreseen from \textsuperscript{47,48,49}Ti(p,2p) reaction. In the event of secondary neutrons were reflected back and hit the \textsuperscript{nat}Ti target, several stable \textsuperscript{47,48,49}Ti atoms were predicted to be produced, whereas short-lived \textsuperscript{51}Ti radioisotope was expected to rise from \textsuperscript{46}Ti(p,d)\textsuperscript{45}Ti reaction. This theoretical data could be useful for future radionuclide production should \textsuperscript{nat}Ti is used as a target window instead of Havar.

1. Introduction

Over the past decades, cyclotrons have been employed to produce radionuclides relevant to nuclear medicines. The well-established radionuclide production includes \textsuperscript{18}F, \textsuperscript{64}Cu [1-4] and many other radionuclides. In cyclotron based-radionuclide production for nuclear medicine applications, particularly when liquid or gas is used as a target, window material separating the cyclotron vacuum chamber and the target influences radionuclide impurities which could eventually contaminate the desired radionuclides. The presence of radionuclide impurities could harm the radiation workers as well as patients please provide what’s kind of harm. Therefore, it is very important to choose the best window material which produces radionuclide impurities when the proton beam passes through the window. The most commonly used window is Havar which comprises of cobalt (42.5 %), chromium (20 %), iron (18.1 %), nickel (13 %), tungsten (2.8 %), molybdenum (2 %), manganese (1.6 %), carbon (0.2 %), beryllium (0.04 %), and addition of some other minor elements [1,2,5]. Meanwhile, the target body is usually made from silver or niobium [1,2,5,6]. Previous investigations reported that various radionuclide impurities as a result of 11-MeV proton bombardment of Havar window included \textsuperscript{56}Co, \textsuperscript{52}Mn, \textsuperscript{60}Co and \textsuperscript{54}Mn [1], whereas other finding highlighted more radionuclide impurities such as \textsuperscript{7}Be,\textsuperscript{48}V, \textsuperscript{51}Cr, \textsuperscript{55}Co, \textsuperscript{57}Co, \textsuperscript{58}Co, \textsuperscript{57}Ni, \textsuperscript{89}Zr, \textsuperscript{92}Nb, \textsuperscript{93}Mo, \textsuperscript{95}Tc and \textsuperscript{96}Tc when 18 MeV protons were employed in the enriched water target and enriched \textsuperscript{93}Nb foil was used as the window [7].
It can be seen from previous investigations that various impurities could be generated when a proton beam hits either window material as well as the target body. Therefore, it is of paramount importance to choose the best materials which contribute to fewer impurities in $^{18}$F production or any other radionuclide production. In this work, we employ the TALYS code to calculate nuclear cross-sections of $^{nat}$Ti used as a window and/or target body. Nuclear reactions as a result of proton irradiation is evaluated and the produced radionuclides are identified from various nuclear reactions, i.e. $^{nat}$Ti(p,n), $^{nat}$Ti(p,2n), $^{nat}$Ti(p,α), $^{nat}$Ti(p,d) and $^{nat}$Ti(n,γ). Both stable and radioactive isotopes predicted as impurities are highlighted. Note that there has been no published work on the theoretical studies on stable and radioactive isotopes produced by proton bombardment of titanium target.

2. Materials and Methods

In this work, the TALYS code [8,9], which can be downloaded from https://tendl.web.psi.ch/tendl_2019/talys.html, was employed to calculate various significant nuclear reactions to obtain nuclear cross-sections of particular reactions, which presumably could generate radioactive and/or stable isotopes. The TALYS code has been widely used in radionuclide production [1,3,10,11]. Five most important nuclear reactions, i.e. $^{nat}$Ti(p,n), $^{nat}$Ti(p,2n), $^{nat}$Ti(p,α), $^{nat}$Ti(p,d) and $^{nat}$Ti(n,γ) were evaluated and the end products, either stable or radioactive isotopes were identified. In this work neutron interaction with $^{nat}$Ti was also studied since they might be reflected back and hit $^{nat}$Ti. In the TALYS code, the nuclear cross-section for an incident particle $p$ and target $t$, $\sigma_{pt}$, was calculated by the compound nucleus formula for a binary cross-section, which is shown in equation (1):

$$\sigma_{pt} = \pi \frac{T_{pt} T_{c}}{k^2 \Sigma c T_{c}} W_{pt}$$ (1)

where $\pi$ is the parity, $k$ is the wavenumber of relative motion, $T$ is the transmission coefficient, $W$ is the width fluctuation correction factor, and $T_{c}$ is the outgoing transmission coefficient. Details on the numerical calculation have been previously discussed elsewhere [8].

In this TALYS calculations, the parameters used as inputs are the incoming particle (proton), the target (natural titanium), proton initial energy (1 MeV), proton final energy (60 MeV). All other parameters such as the wavenumber, transmission coefficient, width of fluctuation correction factor and the ongoing transmission coefficient are commonly used in the TALYS calculations which were set as default. The results of the TALYS calculations are nuclear cross-sections and threshold energy.

3. Results and Discussion

3.1 $^{nat}$Ti(p,n) and $^{nat}$Ti(p,2n) nuclear reactions

Since natural Ti ($^{nat}$Ti) consists of several Ti atoms, namely $^{46}$Ti, $^{47}$Ti, $^{48}$Ti, $^{49}$Ti, and $^{50}$Ti, there might be several isotopes produced when proton irradiated to natural Ti target. As shown in Figure 1, the TALYS calculated data indicates that the cross-sections of individual Ti atoms decrease with decreasing the mass number. Nuclear reaction $^{50}$Ti(p,n) has the highest maximum value of nuclear cross-section (863 mb) compared to the other individual Ti atoms, whereas $^{46}$Ti(p,n) has the lowest maximum value of nuclear cross-section (124 mb).
As can be seen in Table 1, $^{46}$V radionuclide is predictedly generated from $^{46}$Ti(p,n)$^{46}$V nuclear reaction with a threshold energy of 8.83 MeV. Radionuclide $^{46}$V is a positron emitter with a half life of 422.5 milliseconds. Due to the relatively low intensity of gamma energy emitted by $^{46}$V, this radionuclide may not be observed with a relatively poor efficiency gamma spectroscopic system. Another vanadium radionuclide that could be produced by proton bombardment of nat Ti is $^{47}$V, which is due to $^{47}$Ti(p,n)$^{47}$V nuclear reaction with threshold energy of 3.88 MeV. Please provide the formula to calculate threshold energy and why we need to calculate it. From where you know the mode of decay. Radionuclide $^{47}$V decays by emitting positron with a half life of 32.6 minutes. Several gamma rays also follow $^{47}$V decay, though their intensities are relatively low; thus a poor efficiency gamma spectroscopic system may not be able to capture this radionuclide. Note that the threshold energies listed in Table 1 are derived from the TALYS calculation, whereas the other nuclear data such as decay mode, gamma energy and intensity, as well as half-life, are collected from the Tool for Radiation Alarm and Commodity Evaluation (TRACE) application published by IAEA, which can be downloaded via google store.

Table 1. Various isotopes predicted for Ti(p,n) nuclear reactions.

| Isotope | Nuclear reaction | Threshold energy (MeV) | Decay mode | $E_{\gamma}$ in keV (intensity) | Half-life |
|---------|------------------|------------------------|------------|--------------------------------|-----------|
| $^{46}$V | $^{46}$Ti(p,n)$^{46}$V | 8.83 | $\beta^+$ | 4317 (0.0113) 1722 (0.0039) 889 (0.0039) | 422.5 ms |
| $^{47}$V | $^{47}$Ti(p,n)$^{47}$V | 3.88 | $\beta^+$ | 1793 (0.191) 159.8 (0.107) 244.4 (0.094) 1390.4 (0.079) 2163 (0.067) 1549 (0.066) | 32.6 min |
| $^{48}$V | $^{48}$Ti(p,n)$^{48}$V | 5.21 | $\beta^+$ | 983.525 (99.98) 1312.106 (98.2) 944.13 (7.87) 2240.396 (2.33) 928.327 (0.783) | 17.97 min |
| $^{49}$V | $^{49}$Ti(p,n)$^{49}$V | 1.51 | EC | 4.475 (11.8) | 330 d |
Radionuclide $^{48}\text{V}$ may also be formed during proton interaction with $^{nat}\text{Ti}$, which is a result of $^{49}\text{Ti(p,n)}^{48}\text{V}$ nuclear reaction with threshold energy 5.21 MeV. Radionuclide $^{48}\text{V}$ emits positron with a half-life of 17.97 minutes. Since it also emits gamma rays at very high intensities, it could easily recognize by even a poor efficiency spectroscopic system. A previous investigation by Berridge et al [12] reported $^{48}\text{V}$ radionuclide when the proton beam was bombarded to titanium using MC-17 cyclotron. Apart from that, two other vanadium radionuclides, i.e. $^{49}\text{V}$ and $^{50}\text{V}$ are also predicted to occur via $^{49}\text{Ti(p,n)}^{49}\text{V}$ and $^{50}\text{Ti(p,n)}^{50}\text{V}$ nuclear reactions at threshold energies 1.51 and 3.58 MeV respectively. Both of the radionuclides decay through electron capture (EC) with half-lives of 330 days and $1.4 \times 10^{17}$ years. They both may be detectable using a good efficiency spectroscopic gamma system since their gamma-ray emission intensities are relatively high.

As depicted in Figure 2, the TALYS calculated nuclear cross-sections of individual Ti atoms for $(p,2n)$ nuclear reactions are quite similar to that of $(p,n)$ reactions, in which they increase with increasing mass number. Nuclear reaction $^{50}\text{Ti(p,2n)}$ has the highest maximum value of nuclear cross-section (688 mb) compared to the other individual Ti atoms, whereas $^{46}\text{Ti(p,n)}$ has the lowest maximum value of nuclear cross-section (2.42 mb).

As shown in Table 2, vanadium radionuclides, ranging from $^{45}\text{V}$ to $^{49}\text{V}$ are expected to be generated from $^{nat}\text{Ti(p,2n)}$ nuclear reaction with threshold energies ranging from 12.57 to 21.56 MeV, which are higher than the $(p,n)$ reactions. Most produced vanadium radionuclides are positron emitter, except for $^{49}\text{V}$, which decays through electron capture (EC). The half-lives of vanadium radionuclides are relatively short with a maximum half-life of 330 days for $^{49}\text{V}$. Almost all vanadium radionuclides would be very difficult to detect using a gamma-ray spectroscopic system since the intensities of the gamma rays emitted are very low, except for $^{48}\text{V}$, which has very high intensity, i.e. 99.98% for $E_{\gamma} = 983.525$ keV and 98.2% for $E_{\gamma} = 1312.106$ keV. The individual vanadium radionuclides would also be difficult to recognize from their gamma-ray emission at 511 keV (as a result of positron emissions) since they coincide with each other at exactly the same energy.
Table 2. Various isotopes predicted for Ti(p,2n) nuclear reaction.

| Isotope | Nuclear reaction | Threshold energy (MeV) | Decay mode | Eγ in keV (intensity) | Half-life |
|---------|------------------|------------------------|------------|----------------------|-----------|
| 45V 46V | 46 Ti(p,2n) 45 V | 21.56                  | β⁺         | 40.1 (3.5)           | 547 ms   |
| 46V 47V | 47 Ti(p,2n) 46 V | 17.07                  | β⁺         | 4317 (0.0113)        | 422.5 ms |
|        |                  |                        |            | 1722 (0.0039)        |           |
|        |                  |                        |            | 889 (0.0039)         |           |
| 47V 48V | 48 Ti(p,2n) 47 V | 15.66                  | β⁺         | 1793 (0.191)         | 32.6 min  |
|        |                  |                        |            | 159.8 (0.107)        |           |
|        |                  |                        |            | 244.4 (0.094)        |           |
|        |                  |                        |            | 1390.4 (0.079)       |           |
|        |                  |                        |            | 2163 (0.067)         |           |
|        |                  |                        |            | 1549 (0.066)         |           |
| 48V 49V | 49 Ti(p,2n) 48 V | 13.21                  | β⁺         | 983.525 (99.98)      | 17.97 min |
|        |                  |                        |            | 1312.106 (98.2)      |           |
|        |                  |                        |            | 944.13 (7.87)        |           |
|        |                  |                        |            | 2240.396 (2.33)      |           |
|        |                  |                        |            | 928.327 (0.783)      |           |
| 49V 50V | 50 Ti(p,2n) 49 V | 12.57                  | EC         | -                    | 330 d    |

3.2 natTi(p,α) and natTi(p,d) nuclear reactions

When proton beam impinges to natTi window or target body, alpha particles may be created followed by radioactive isotopes or stable isotopes. Based on the TALYS-calculated nuclear cross-sections natTi(p,α) as shown in Figure 3, 47Ti(p,α) has the highest maximum value of nuclear cross-section (84.8 mb), whereas 50Ti(p,α) has the lowest maximum value of nuclear cross-section (17.6 mb).

![Figure 3. TALYS calculated nuclear cross-sections of individual Ti atoms for (p,α) nuclear reactions.](image)

As listed in Table 3, scandium isotopes are produced from natTi(p,α) nuclear reactions, in which stable 45Sc is generated from 48Ti(p, α)45Sc reaction with threshold energy of 2.61 MeV. The other scandium isotopes produced from natTi(p,α) reactions are radioactive which emit positrons with relatively short half-lives ranging from 3.89 hours for 43Sc to 83.79 days for 46Sc. All scandium radionuclides should be detectable even with a poor efficiency gamma-ray spectroscopic system since
they have relatively high gamma-ray intensities. The threshold energies are ranging from as low as 1.98 MeV for $^{46}\text{Sc}$ to 3.14 MeV for $^{43}\text{Sc}$.

### Table 3. Various isotopes predicted for Ti(p,α) nuclear reactions.

| Isotope | Nuclear reaction | Threshold energy (MeV) | Decay mode | $E_\gamma$ in keV (intensity) | Half-life |
|---------|------------------|------------------------|------------|-------------------------------|-----------|
| $^{43}\text{Sc}$ | $^{46}\text{Ti(p,α)}^{43}\text{Sc}$ | 3.14 | $\beta^+$ | 372.9 (22.5) | 3.89 h |
| $^{44}\text{Sc}$ | $^{47}\text{Ti(p,α)}^{44}\text{Sc}$ | 2.30 | $\beta^+$ | 1157.02 (99.9) | 3.97 h |
| $^{45}\text{Sc}$ | $^{48}\text{Ti(p,α)}^{45}\text{Sc}$ | 2.61 | stable | 1120.545 (99.99) | - |
| $^{46}\text{Sc}$ | $^{49}\text{Ti(p,α)}^{46}\text{Sc}$ | 1.98 | $\beta^+$ | 889.277 (99.98) | 83.79 d |
| $^{47}\text{Sc}$ | $^{50}\text{Ti(p,α)}^{47}\text{Sc}$ | 2.28 | $\beta^+$ | 159.381 (68.3) | 3.349 d |

There is also a great possibility of deuteron production when protons are irradiated to the $^{nat}\text{Ti}$ target. The TALYS calculated nuclear cross-sections of individual natural titanium atoms are shown in Figure 4, which indicate that $^{47}\text{Ti(p, d)}$ has the highest maximum value of nuclear cross-section (37.3 mb), whereas $^{46}\text{Ti(p, d)}$ has the lowest maximum value of nuclear cross-section (25.7 mb).

![Figure 4. TALYS-calculated nuclear cross-sections of individual Ti atom for (p,d) nuclear reactions.](image)

Assummarized in Table 4, titanium isotopes are produced from $^{nat}\text{Ti(p,d)}$ reactions. Most of the generated titanium isotopes are stable, whereas only one isotope is radioactive, i.e. $^{45}\text{Ti}$ which is formed from $^{46}\text{Ti(p, d)}^{45}\text{Ti}$ nuclear reaction. Radionuclide $^{45}\text{Ti}$ emits positron with half-life of 184.8 minutes and threshold energy of 11.21 MeV. The gamma-ray intensities emitted by $^{45}\text{Ti}$ radionuclide are very low, which makes it very difficult to detect particularly with a poor efficiency gamma-ray spectroscopic system. Nevertheless, it might be recognized from gamma-ray emitted at 511 keV (due
to positron emission), though it would coincide with other positron emitters resulted from proton bombardment of natTi target.

Table 4. Various isotopes predicted for Ti(p,d) nuclear reactions.

| Isotope | Nuclear reaction | Threshold energy (MeV) | Decay mode | Eγ in keV (intensity) | Half-life |
|---------|------------------|------------------------|------------|-----------------------|-----------|
| 45Ti    | 46Ti(p,d)45Ti     | 11.21                  | β⁺         | 719.6 (0.154)         | 184.8 min |
|         |                   |                        |            | 1408.1 (0.085)        |           |
|         |                   |                        |            | 1660.9 (0.041)        |           |
|         |                   |                        |            | 425 (0.0137)          |           |
|         |                   |                        |            | 1236.5 (0.0118)       |           |
| 46Ti    | 47Ti(p,d)46Ti     | 6.80                   | stable     | -                     | -         |
| 47Ti    | 48Ti(p,d)47Ti     | 9.60                   | stable     | -                     | -         |
| 48Ti    | 49Ti(p,d)48Ti     | 6.04                   | stable     | -                     | -         |
| 49Ti    | 50Ti(p,d)49Ti     | 8.89                   | stable     | -                     | -         |

3.3 Isotopes produced from secondary neutrons

Following proton bombardment of natTi target, secondary protons are produced, particularly from (p,n) and (p,2n) reactions, and their energies are distributed from cold neutrons to fast neutrons. These neutrons could potentially hit surrounding materials including the natTi target. Based on the TALYS calculated nuclear cross-sections of individual natural Ti targets, the (n,γ) cross-sections are very high as indicated in Figure 5. The highest nuclear cross-section is due to 46Ti(n,γ) reaction, which is 5.37x10⁴ mb, whereas the lowest one is from 50Ti(n,γ) reaction, which is 1.47x10⁴ mb.

Figure 5. TALYS-calculated nuclear cross-sections of individual Ti atoms for (n,γ) nuclear reactions

As listed in Table 5, titanium isotopes could be produced following proton bombardment of natTi target via (n,γ) reaction. All stable titanium isotopes ranging from 47Ti to 50Ti are generated with 0
MeV threshold energies. The only titanium radionuclide as a result of \((n,\gamma)\) reaction is \(^{51}\)Ti which is produced from \(^{50}\)Ti\((n,\gamma)^{51}\)Ti nuclear reaction. Radionuclide \(^{51}\)Ti decays by beta emission with a half-life of 5.76 minutes. Radionuclide \(^{51}\)Ti could be captured using a gamma spectroscopy system since its intensity is quite high (68.3%).

| Isotope | Nuclear reaction | Threshold energy (MeV) | Decay mode | \(E_\gamma\) in keV (intensity) | Half-life |
|---------|------------------|------------------------|------------|---------------------------------|-----------|
| \(^{47}\)Ti | \(^{46}\)Ti\((n,\gamma)^{47}\)Ti | 0 | stable | - | - |
| \(^{48}\)Ti | \(^{47}\)Ti\((n,\gamma)^{48}\)Ti | 0 | stable | - | - |
| \(^{49}\)Ti | \(^{48}\)Ti\((n,\gamma)^{49}\)Ti | 0 | stable | - | - |
| \(^{50}\)Ti | \(^{49}\)Ti\((n,\gamma)^{50}\)Ti | 0 | stable | - | - |
| \(^{51}\)Ti | \(^{50}\)Ti\((n,\gamma)^{51}\)Ti | 0 | \(\beta^-\) | 159.381 (0.683) | 5.76 min |

4. Conclusion

Natural titanium (\(^{nat}\)Ti) interaction with proton beam is, for the first time, comprehensively studied from their possible nuclear interactions with incoming proton using the TALYS code. The TALYS code has been employed to calculate nuclear cross-sections with \(^{nat}\)Ti from five different nuclear reactions, i.e. \(^{nat}\)Ti\((p,n)\), \(^{nat}\)Ti\((p,2n)\), \(^{nat}\)Ti\((p,\alpha)\), \(^{nat}\)Ti\((p,d)\) and \(^{nat}\)Ti\((n,\gamma)\). Based on the TALYS-calculated results, vanadium (V) radionuclides, e.g. \(^{46,47,48,49,50}\)V could be produced from \(^{nat}\)Ti\((p,n)\), whereas \(^{45,46,47,48,49}\)V could be generated from \(^{nat}\)Ti\((p,2n)\) nuclear reaction. Interaction between proton beam and \(^{nat}\)Ti could also possibly produce \(^{43,44,46,47}\)Sc radionuclides and stable \(^{45}\)Sc isotope which are due to \(^{nat}\)Ti\((p,\alpha)\) reaction. For \(^{nat}\)Ti\((p,d)\) reaction, \(^{45}\)Ti radionuclide is expected to be generated, while other stable \(^{46,47,48,49,50}\)Ti atoms could be created as a result of this nuclear reaction. Furthermore, stable isotopes, i.e. \(^{47,48,49,50}\)Ti and radionuclide \(^{51}\)Ti might be produced from \(^{nat}\)Ti\((n,\gamma)\) reaction.

5. References

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