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Transmutation of ^{129}I Containing Nuclear Waste by Proton Bombardment

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Transmutation of ^{129}I Containing Nuclear Waste by Proton Bombardment

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Abstract. As a long-lived nuclear waste, iodine (^{129}I) which is primarily generated from nuclear fission of uranium or plutonium is considered harmful to human and environment. Therefore, proper steps are required to treat the radioactive isotope. In this work, we propose transmutation of I-129 isotope using cyclotron-based proton bombardment. The TALYS code was employed to calculate nuclear cross-sections of various nuclear reactions possible to transmute I-129 into short-lived radionuclides or stable isotopes. Twelve different nuclear reactions, namely (p,n), (p,2n), (p,3n), (p,np), (p,2np), (p,α), (p,d), (p,2p), (p,nα), (p,γ), (p,^{3}He), and (p,t), which have significant cross-sections (greater than 1 mb) were analyzed. Based on the nuclear reactions, there were 6 possible stable isotopes produced from proton bombardment of ^{129}I, such as ^{129}Xe, ^{128}Xe, ^{127}I, ^{126}Te, ^{125}Te and ^{130}Xe. Short-lived radionuclides such as ^{129m}Xe (half life = 8.88 days), ^{127}Xe (half life = 36.35 days), ^{127m}Xe (half life = 62.9 seconds), ^{128}I (half life = 24.99 minutes), ^{125m}Te (half life = 57.40 days), ^{127}Te (half life = 9.35 hours) and ^{127m}Te (half life = 106.1 days) could also be possibly produced from the proton irradiation of ^{129}I nuclear waste. To sum up, the longest radionuclide could be generated from proton bombardment of ^{129}I nuclear waste is ^{127m}Te (half life = 106.1 days), which is a lot shorter than ^{129}I (half life = 1.57×10^7 years). This theoretical study indicates that transmutation of ^{129}I nuclear waste by proton bombardment into short-lived radionuclides is greatly feasible. Current available cyclotrons in Indonesia may be employed to help transmute ^{129}I into short-lived radionuclides or stable isotopes. This theoretical study can be used as a reference for future ^{129}I nuclear waste if proton beams are employed in the transmutation.

1. Introduction

Nuclear reactor is a source of neutrons which can be used to produce various medical radioisotopes such as technetium-99m (^{99m}Tc) [1], lutetium-177 (^{177}Lu) [2-3], samarium-153 (^{153}Sm) [4] and many others which are employed in both diagnostic and therapeutic procedures in nuclear medicine. While nuclear reactor can be very useful for radioisotope production it is, on the other hand, a source of nuclear waste. Nuclear fission occurs when neutrons interact with uranium or plutonium fuel. This fission reaction produces various long-lived radioactive isotopes, including technetium-99 (^{99}Tc), plutonium-242 (^{242}Pu), neptunium-237 (^{237}Np), iodine-129 (^{129}I) [5], tin-126 (^{126}Sn), selenium-79 (^{79}Se), zirconium-93 (^{93}Zr), caesium-135 (^{135}Cs) and palladium-107 (^{107}Pd). Therefore, maximum precaution should be implemented to the long-lived radioactive isotopes.

Origen 2 code-based theoretical research on transmutation of nuclear waste using thermal and fast neutrons has been reported elsewhere which found that transmutation factor was reduced to 0.8–0.5 after 1000 years when the spent nuclear fuel was irradiated with thermal neutrons [6]. Accelerator driven neutron source has also been studied which suggested production of thermal and fast neutrons for nuclear waste transmutation [7-9]. Irani et al [10] suggested the use of laser for transmutation of ^{126}Sn into short-lived nuclear medicine ^{125}Sn. Other important work on nuclear waste transmutation has reported proton beams as feasible source for nuclear waste transmutation [11-12], while fusion-driven system has also been highlighted elsewhere [13-15].

As mentioned above, iodine-129 (^{129}I) is one of the long-lived radioactive isotopes found in nuclear waste and is primarily produced from the fission of uranium or plutonium in nuclear reactor. It emits beta particle with average energy of 40 keV and has half life of $1.57x10^7$ years. Due to its very long half life and beta emission, it is considered harmful to radiation workers; thus nuclear waste containing I-129 should be properly treated. This work aims at theoretically studying the possibility of transmutation of I-29 nuclear waste into short-lived and stable isotopes by proton bombardment. The feasible paths are analyzed from their nuclear cross-sections should proton beams be employed as the incident particle. In addition, the end-products of the interactions are also highlighted and discussed from their half life. The proton sources are expected from medium and high energy cyclotron commercially available worldwide. To the best of the author knowledge, there has been no published reference on transmutation of ^{129}I using proton beam.

2. Materials and Methods

In this present theoretical study, pure radioactive waste containing ^{129}I radioisotope only was simulated as a target, while proton beams of up to 200 MeV were employed as the incident particles. Nuclear reactions as a result of the proton bombardments were then analyzed from their nuclear cross-sections using the TALYS code [16-17]. In this work, the Talys code version 1.9 was used to compute nuclear cross-sections of several reactions. Twelve (12) different nuclear reactions, namely (p,n), (p,2n), (p,g), (p,d), (p,α), (p,t), (p,np), (p,2p), (p,p2n), (p,nα) and (p,He-3) which have significant cross-sections (greater than 1 mb) were analyzed and the resulting isotopes were predicted. The TALYS code has been widely used in the studies of various radioisotope production [18-26], though it has never been used to study transmutation of nuclear waste. Experimental research on ^{57}Co and ^{57}Ni productions have also been reportedly used the TALYS code [27].

3. Results and Discussion

3.1 Isotopes produced from (p,n), (p,2n), (p,3n), (p,np) and (p,2np) nuclear reactions

Based on the TALYS calculated nuclear cross-sections of (p,n), (p,2n), (p,3n), (p,np) and (p,2np) nuclear reactions between proton and ^{129}I, (p,2n) reaction has the highest cross-section of up to 1070 mb whereas (p,3n) has the second highest cross-section (936 mb) as shown in Figure 1. The three other nuclear reactions, i.e. (p,n), (p,np) and (p,2np) have cross-sections of less than 310 mb. Proton bombardment of ^{129}I via (p,n) nuclear reaction results in production of stable isotope ^{129}Xe. The threshold energy for $^{129}I(p,n)^{129}Xe$ is 0.63 MeV. Another possible isotope production from (p,n) reaction is formation of short-live radionuclide ^{129m}Xe (half life = 8.88 days) via $^{129}I(p,n)^{129m}Xe$ wich decays through isomeric transition (IT).

Proton irradiation of ^{129}I through (p,2n) nuclear reaction could generate stable isotope ^{128}Xe with threshold energy of 7.56 MeV for $^{129}I(p,2n)^{128}Xe$ reaction. There are two possible radionuclides produced from (p,3n) nuclear reaction, i.e. ^{127}Xe via $^{129}I(p,3n)^{127}Xe$ and ^{127m}Xe through $^{129}I(p,3n)^{127m}Xe$. Radionuclide ^{127}Xe decays through β^+ emission with half life of 36.35 days, whereas ^{127m}Xe radionuclide decays via isomeric transition (IT) with half life of 62.9 seconds.

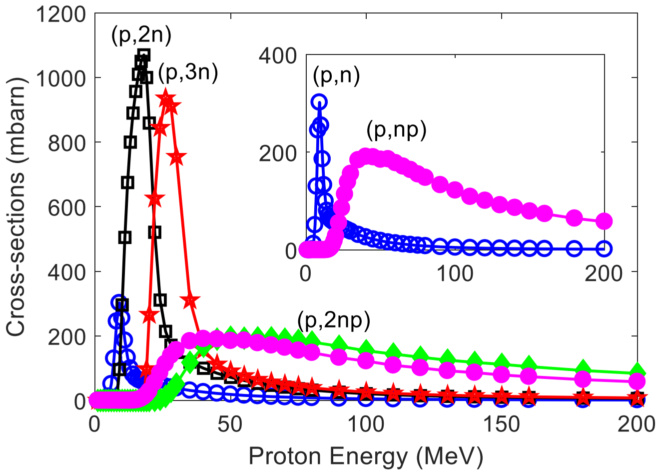

Figure 1. TALYS calculated nuclear cross-section of (p,n), (p,2n), (p,3n), (p,np) and (p,2np)

Proton bombardment of ^{129}I via (p,np) nuclear reaction results in formation of short-lived ^{128}I radionuclide (half life = 24.99 minutes) which decays by beta emission. The threshold energy for ^{129}I(p,np)^{128}I is 8.91 MeV. On the other hand, proton irradiation of ^{129}I through (p,2np) nuclear reaction could produce stable isotope ^{127}I with threshold energy 15.8 MeV. The complete nuclear data for (p,n), (p,2n), (p,3n), (p,np) and (p,2np) nuclear reactions are summarized in Table 1.

Table 1. Possible isotopes produced as a result of proton-bombarded ^{129}I for (p,n), (p,2n), (p,3n), (p,np) and (p,2np) nuclear reactions

Isotope	Nuclear Reaction	Threshold energy (MeV)	Decay mode	Half life
^{129}Xe	^{129}I(p,n)^{129}Xe ^{129}I(p,n)	0.63	Stable	-
^{129m}Xe	^{129m}Xe	0.63	IT	8.88 d
^{128}Xe	^{129}I(p,2n)^{128}Xe	7.56	Stable	-
^{127}Xe	^{129}I(p,3n)^{127}Xe	17.2	β^{+}	36.35 d
^{127m}Xe ^{128}I	^{128}Te(p,3n)^{127m}Xe	17.2	IT	62.9 s
^{127}I	^{129}I(p,np)^{128}I	8.91	β^{-}	24.99 m
	^{129}I(p,2np)^{127}I	15.8	stable	-

3.2 Isotopes produced from (p,α), (p,d), (p,2p), (p,nα), (p,γ), (p,^{3}He), and (p,t) nuclear reactions

Based on the TALYS calculated nuclear cross-sections of (p,α), (p,d), (p,2p), (p,nα), (p,γ), (p,^{3}He), and (p,t) nuclear reactions between proton and ^{129}I, (p,d) reaction has the highest cross-section of up to 26.8 mb whereas (p,2p) has the second highest cross-section (23.3 mb) as shown in Figure 2. The five other nuclear reactions, i.e. (p,α), (p,nα), (p,γ), (p,^{3}He), and (p,t) have cross-sections of less than 15 mb.

Proton bombardment of ^{129}I via (p,α) nuclear reaction results in production of stable isotope ^{126}Te. The threshold energy for ^{129}I(p,n) ^{126}Te reaction is 0 MeV. When proton is irradiated to ^{129}I, then another possible radionuclide generation is ^{128}I which decays by beta emission at half life of 24.99 minutes via ^{129}I(p,d)^{128}I nuclear reaction. The threshold energy for ^{129}I(p,d)^{128}I reaction is 6.67 MeV.
While very long half life ^{128}Te radionuclide (half life = 7.7x10^{24} y) maybe produced from ^{129}I(p,2p)^{128}Te nuclear reaction, the resulting ^{128}Te can simultaneously transmute into short-lived ^{128}I radionuclide (half life = 24.99 minutes) by further proton bombardment via ^{128}Te(p,n)^{128}I reaction. The threshold energy for ^{128}Te(p,n)^{128}Ireaction itself is 2.08 MeV. It should be noted that Te-128 separation from the nuclear waste is not required since once it is generated, the incoming proton beam directly irradiate it to form ^{128}I radionuclide.

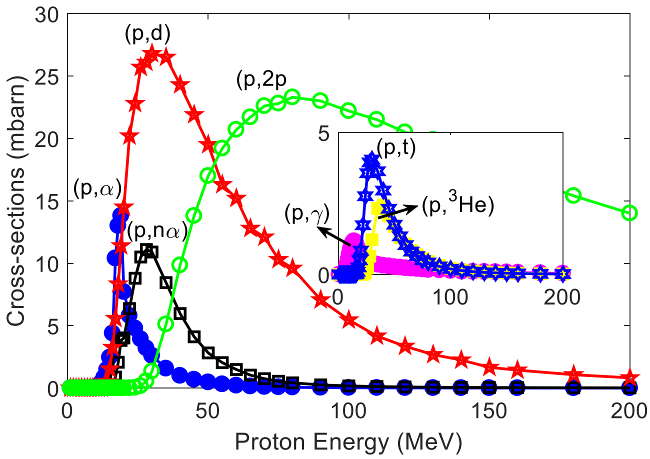

Figure 2. TALYS calculated nuclear cross-section of (p,α), (p,d), (p,2p), (p,nα), (p,γ), (p,^{3}He), and (p,t)

Proton irradiation of ^{129}I through (p,nα) nuclear reaction could possibly generate two isotopes, either stable isotope ^{125}Te with threshold energy of 2.71 MeV for ^{129}I(p,2n)^{128}Xe reaction or radionuclide ^{125m}Te. Radionuclide ^{125m}Te decays via isomeric transition (IT) with half life of 57.40 days. For proton bombardment of ^{129}I through (p,γ) nuclear reaction, stable isotope ^{130}Xe maybe produced. Again, there are two possible radionuclides produced from (p,^{3}He) nuclear reaction, i.e. ^{127}Te via ^{129}I(p,3n) ^{127}Te reaction and ^{127m}Te through ^{129}I(p,^{3}He) ^{127m}Te reaction. Radionuclide ^{127}Te decays through β^- emission with half life of 9.35 hours, whereas ^{127m}Te radionuclide decays via isomeric transition (IT) with half life of 106.1 days. Furthermore, stable isotope ^{127}I could possibly be generated as a result of ^{129}I (p,t) ^{127}I nuclear reaction with threshold energy of 2.08 MeV. The complete nuclear data for (p,α), (p,d), (p,2p), (p,nα), (p,γ), (p,^{3}He), and (p,t) nuclear reactions are summarized in Table 2.

Table 2. Possible produced isotopes as a result of proton-bombarded ^{129}I for (p,α), (p,d), (p,2p), (p,nα), (p,γ), (p,^{3}He), and (p,t) nuclear reactions

Isotope	Nuclear Reaction	Threshold energy (MeV)	Decay mode	Half life
^{126}Te	^{129}I(p,α)^{126}Te	0	Stable	-
^{128}I	^{129}I(p,d)^{128}I	6.67	β^-	24.99 m
^{128}Te	^{129}I(p,2p)^{128}Te	6.86	2β^-	7.7x10^{24} y
^{128}I	^{128}Te(p,n)^{128}I	2.08	β^-	24.99 m
^{125}Te	^{129}I(p,nα)^{125}Te	2.71	Stable	-
^{125m}Te	^{129}I(p,nα)^{125m}Te	2.71	IT	57.40 d
^{130}Xe	^{129}I(p,γ)^{130}Xe	0	Stable	-
^{127}Te	^{128}Te(p,^{3}He)^{127}Te	7.93	β^-	9.35 h
^{127m}Te	^{129}I(p,^{3}He)^{127m}Te	7.93	IT	106.1 d
^{127}I	^{129}I (p,t)^{127}I	2.08	stable	-

Currently, there are 3 available cyclotrons in Indonesia which may be employed for transmutation of ^{129}I nuclear waste. The three cyclotrons are 9 MeV cyclotron in Gading Pluit Hospital, Jakarta, 11 MeV cyclotron in Dharmais Cancer Hospital and 18 MeV cyclotron in Siloam Hospital, Jakarta. Based on the energy characteristics of protons generated from the three cyclotrons, it is expected that the three cyclotrons would be capable of slightly different radionuclides, depending on the accelerated proton energy and the threshold energy. As discussed earlier in Table 1 and Table 2, nearly all isotopes are possible to be produced using the three cyclotrons, except for ^{127}Xe and ^{127m}Xe which can be generated using 18 MeV cyclotron in Siloam Hospital since their threshold energies are 17.2 MeV.

4. Conclusion

Possible transmutation of nuclear waste containing ^{129}I has been theoretically studied using proton beam as incident particle. Possible nuclear reactions and produced isotopes was determined from the TALYS 2017 calculated nuclear cross-sections. Twelve different nuclear reactions, i.e. (p,n), (p,2n), (p,3n), (p,g), (p,d), (p,α), (p,t), (p,np), (p,2p), (p,2np), (p,nα) and (p,^{3}He) which have significant cross-sections (greater than 1 mb) were analyzed and the produced stable isotopes and radionuclides were highlighted. Based on this study, there were 6 possible stable isotopes produced from proton bombardment of ^{129}I, i.e. ^{129}Xe, ^{128}Xe, ^{127}I, ^{126}Te, ^{125}Te and ^{130}Xe. In addition, there were 7 short-lived radionuclides such as ^{129m}Xe (half life = 8.88 days), ^{127}Xe (half life = 36.35 days), ^{127m}Xe (half life = 62.9 seconds), ^{128}I (half life = 24.99 minutes), ^{125m}Te (half life = 57.40 days), ^{127}Te (half life = 9.35 hours) and ^{127m}Te (half life = 106.1 days) could also be possibly produced from the proton irradiation of ^{129}I nuclear waste. It is clear that the longest radionuclide could be generated from proton bombardment of ^{129}I nuclear waste is ^{127m}Te (half life = 106.1 days), which is a lot shorter than ^{129}I (half life = 1.57x10^7 years). In addition, current available cyclotrons (9, 11 and 18 MeV cyclotrons) in Indonesia may be employed to help transmute ^{129}I into short-lived radionuclides or stable isotopes.

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