Comparison of 192Os(p,n)192Ir and 192Os(d,2n)192Ir Nuclear Reactions for 192Ir Production

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Comparison of ^{192}Os(p,n)^{192}Ir and ^{192}Os(d,2n)^{192}Ir Nuclear Reactions for ^{192}Ir Production

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ABSTRACT

Iridium-192 (^{192}Ir) is a radionuclide currently suggested for brachyteraphy. One of the methods employed to produce high purity ^{192}Ir is by irradiation of Osmium-192 (^{192}Os) target using cyclotron. The success of ^{192}Ir radionuclide production in cyclotrons requires deep understanding of irradiation parameters, including particle energy, target preparation and thickness, particle beam curent and irradiation time. Therefore, theoretical calculations of the ^{192}Ir radioactivity yields should be carried out as a preliminary measure for more efficient ^{192}Ir production. In this study, ^{192}Ir production was simulated using the SRIM 2013 program to determine the optimum target thickness while the nuclear cross-section data were extracted from TENDL 2017. Two nuclear reactions for ^{192}Ir production yield calculations were compared, i.e., ^{192}Os(p,n)^{192}Ir and ^{192}Os(d,2n)^{192}Ir. The radioactivity yields for ^{192}Os(p,n)^{192}Ir nuclear reaction was found to be lower than ^{192}Os(d,2n)^{192}Ir reaction. For proton and deuteron energy of 30 MeV, the maximum radioactivity yield was 6.79 GBq for ^{192}Os(p,n)^{192}Ir and 26.14 GBq for ^{192}Os(d,2n)^{192}Ir. Several radionuclide impurities such as ^{191m}Ir, ^{190}Ir, ^{191}Os and ^{189}Re were predicted to be generated during ^{192}Os(p,n)^{192}Ir reaction for proton incident energy between 1 and 30 MeV; meanwhile, ^{192}Ir, ^{191m}Ir, ^{193}Os, ^{193m}Ir, ^{192m}Os and ^{191}Os radionuclides were expected to contaminate during ^{192}Os(d,2n)^{192}Ir reaction for deuteron energy between 1 and 30 MeV. Results of this study can be used as a reference for future ^{192}Ir radionuclide production when proton or deuteron beams are considered to be employed.

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INTRODUCTION

Iridium-192 (^{192}Ir) is a radionuclide currently suggested to be used in radiotherapy by brachytherapy method. This radionuclide has a half-life of 73.83 days and emits beta radiation with a maximum energy of 675 keV and gamma radiation with energy of 317 keV (the highest intensity is 82.8 %). Due to its extensive use in medical application, ^{192}Ir has been artificially produced in nuclear reactors using ^{191}Ir(n,γ)^{192}Ir reaction through irradiation of Na_2IrCl_6 and iridium wire. However, nuclear reactor-based ^{191}Ir production generates low specific activity of ^{192}Ir [1-3].

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One method that can be used to produce ^{192}Ir with high purity is by irradiation of Osmium-192 (^{192}Os) target using cyclotron [3]. The ^{192}Os target material that is bombarded with high-energy charged particles (protons or deuterons) will produce ^{192}Ir radionuclide through ^{192}Os(p,n)^{192}Ir and ^{192}Os(d,2n)^{192}Ir reactions. Cyclotron has been widely used to produced medical radionuclides, and recently it has been suggested for the production of some radionuclides such as ^{47}Sc [4], ^{68}Ga [5-10], ^{64}Cu [11], ^{86}Y [12-14] and ^{99m}Tc [15-18].

During interactions betwen charged particles and the target material, both protons or deuterons will hit the target atoms, causing the particles to lose their energy. The particles will then slow down and eventually stop after reaching a certain range. When the incident particles interact with the target atomic nucleus, there will be absorption of particles followed by the release of nucleon particles from the target atomic nucleus and will finally produce a new unstable nucleus (radionuclide).

The success of radionuclide production in cyclotrons requires deep understanding of irradiation parameters including proton energy, target preparation, proton beam currents and irradiation time [19]. Direct experiments for obtaining these parameters are time and cost consuming, assuming the expensive cost of cyclotron operation, especially if repeated experiments are required to obtain the right parameter values. Therefore, preliminary studies of radionuclide production by theoretical simulations can be considered for better efficiency. This work aimed to calculate the ranges of protons and deuterons in ^{192}Os targets in the energy range between 1 and 30 MeV using the SRIM 2013 code. The ranges were then used as the recommended target thickness for ^{192}Ir production. The nuclear cross-sections were determined from TENDL 2017.

The distribution of charged particles such as protons and deutrons in the ^{192}Os target material can be obtained from particles stopping power dan particles range, which can be calculated using the Stopping and Range of Ion in Matter (SRIM) program [20]. In SRIM code, stopping power is defined as the energy needed to slow down the proton particles bombarded during their interaction with the material at a certain distance, and the total ion stop distance is called the range. From the data of particle stopping power and cross section, the yield value of radiactivity can be calculated. Meanwhile, the particle range can be used to determine the target thickness recommendations.

This work dealt with the calculation of the ranges and stopping powers of protons and deuterons in ^{192}Os targets in the energy range between 1 and 30 MeV using the SRIM 2013 code. The ranges can then be used as the recommended target thickness for ^{192}Ir production. The nuclear cross-sections was determined from TENDL 2017, in which both stopping powers and nuclear cross-sections were then used to calculate the ^{192}Ir radioactivity yield. Since the threshold energy for ^{192}Os(p,n)^{192}Ir nuclear reaction is 1.8 MeV and the excitation function saturates at 30 MeV, the best approach for the calculations is, therefore, between 1 and 30 MeV. Also, the currently available cyclotrons in Indonesia can only accelerate particles up to 30 MeV, thus in this study 30 MeV is chosen as the maximum energy.

THEORY/CALCULATION

The stopping power of charged particles in matter

The linear stopping power of a charged particle (S) that moves through a certain material is physically defined as the energy loss (dE) of a particle that moves across a distance (dx) [20]. This stopping power is also referred to as the rate of energy loss. During the accelerated movement and collision in the target material, the particles will interact with the target's atoms and lose their energy, both energy loss as a result of interaction with the target atomic nucleus (called as nuclear energy loss, Sn) and energy loss as a result of interaction with target atomic electrons (called as electronic energy loss, Se). In total, the energy loss rate (linear stopping power) can be formulated in equation (1) [20].

$$S = -\frac{dE}{dx} = -N(S_n + S_e) \qquad (1)$$

Where N is the density of matter, dE is the energy loss, and dx is the distance traveled by the particle. Meanwhile, Sn and Se are empirically defined as equations (2) and (3).

$$S_n = N \int_{T_d}^{E} T d\sigma \qquad (2)$$

$$S_e = \frac{2\pi Z_{eff} Z_2}{\beta^2} N m_e {r_e}^2 [\ln\left(\frac{2 m_e \beta^2 \gamma^2 T_{maks}}{{I_{av}}^2}\right) - 2\beta^2] \qquad (3)$$

In this case, T is the kinetic energy of particles, σ is a differential particle latitude, Z_{eff} is the effective atomic number of the projectile particle, Z_2 is the target atomic number, m_e is the silent mass of the electron, r_e is the radius of the electron path, I_{av} is a flat ionization potential mean, and β is the relative velocity of the particles [20].

Range of charged particles in matter

At the end of the interaction, charged particles bombarded on a target will stop after a certain range of distance (R), which is defined as the total distance traveled by the particles in the target, the calculation of which encompasses the distance from the particles enter the material until they completely stop at the atomic grid target. This range can be formulated as:

$$R(E) = \int_0^E \left(\frac{dE}{dx}\right)^{-1} dE \approx \sum_0^E \left(\frac{dE}{dx}\right)^{-1} \Delta E \qquad (4)$$

In this case, R is the range of charged particles in matter, E is the energy of the charged particle, and x is the distance traveled by the particles.

Radionuclide yield

Yield is the number of radionuclides formed from a nuclear reaction resulting from bombardment of charged particles. Nuclear reactions are likely to occur when high-energy particles such as a 10 MeV proton beam are bombarded on a target, where, during irradiation and at the end of irradiation, some radioactive isotopes are eventually produced. The yield value (Y) for each radioisotope of nuclear particles produced depends not only on the nuclear cross section of a particular energy σ(E) but also on the stopping power (dE/dx) and several other parameters, as shown in the following equation [20].

$$Y = \varphi(1 - e^{-\lambda t})\frac{N_A}{M}\int_{E_i}^{E_{th}}\left[\frac{\sigma(E)}{\frac{1}{\rho}\frac{d(E)}{dx}}\right]dE \qquad (5)$$

Where φ is the number of time-charged particles, λ is the radioisotope decay constant produced, t is the duration of irradiation, N_A is Avogadro number, ρ and M is the mass density and atomic mass of each target respectively, E_i is the initial energy of incident particles, and E_{th} is the threshold energy of the reaction.

SRIM

SRIM or the stopping and range of ions in matter is a software package that has many calculation features for ion movement in matter. The SRIM program can be used to calculate the stopping power and range of particles or ions with energies of 10 eV/amu up to 2 GeV/amu in the material by applying collision quantum mechanical theory between ions and atoms. In this case the ion is a moving (accelerating) particle while an atom is the target material that is passed by an ion. SRIM can perform a quick calculation that results in a range of stopping power tables and straggling distributions for various ions at various energies in various target elements. More complicated calculations were included the calculations, for targets with complex multilayer configurations.

MATERIALS AND METHODS

The theoretical studies of ^{192}Ir radioisotope production based on ^{192}Os(p,n)^{192}Ir and ^{192}Os(d,2n)^{192}Ir nuclear reactions using the SRIM codes [20] and TENDL 2017 [21] were carried out from October to December 2018 at the Radiation and Medical Physics Laboratory, Faculty of Science and Mathematics, Diponegoro University. The target of interest was ^{192}Os (100 % purity) while the incident particle beams were protons and deuterons. The proton and deuteron energies were varied between 1 and 30 MeV.

The SRIM 2013 code was used to calculate the stopping power and ion range in the ^{192}Os targets, whereas the cross-section data were obtained from TENDL 2017 [21]. In addition, Microsoft Excel was employed for data processing and radioactivity yield calculations using mathematical equation (5). The SRIM 2013 and TENDL 2017 have been previously used to study several radioisotopes production [22,23].

RESULTS AND DISCUSSION

Recommended osmium-192 target thickness

Based on the SRIM 2013 calculation results, it can be seen in Fig. 1(a) and 1(b) that the ranges of protons and deuterons in ^{192}Os targets are strongly dependent on the energy of the incoming particles. The greater the particle energy the deeper the range of the particles in the target material. In addition, the type of target material and particle also influences the range of particles in the target material.

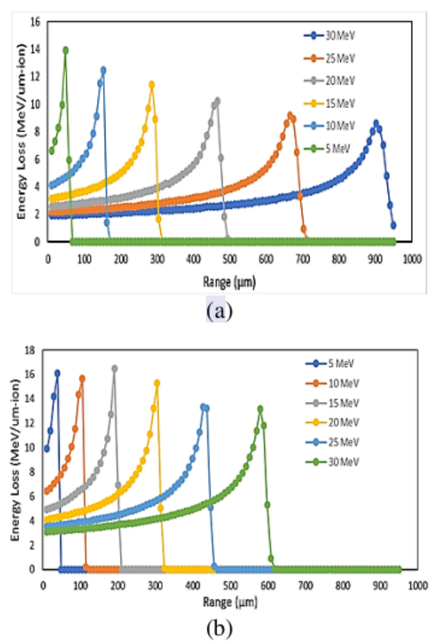

Fig. 1. The relations between energy loss and range of incident particles (a) Proton (b) Deuteron, in ^{192}Os target.

Overall, the stopping power distribution pattern of the ranges of protons and deuterons in the energy range between 5 MeV and 30 MeV in ^{192}Os target is relatively similar, as can be seen in Fig. 1(a) and 1(b). The energy loss or stopping power tends to increase with increasing range before it drops significantly after reaching a certain peak value (brag peak). On the other hand, energy loss or stopping power values decrease with the increasing particle energy.

The target thickness can be determined from the particle range. Table 1 indicates the recommended target thickness of ^{192}Os target for various incident particle energies when the target is bombarded with either protons or deuterons. For instance, for 11 MeV protons incident on ^{192}Os target, the target thickness should be 172.44 μm, whereas when deuterons are employed in the bombardment, the target should be thinner, i.e., 117.52 μm. Overall, thicker ^{192}Os target should be used for irradiation with protons.

Table 1. Ranges of protons and deuterons at different energies

Energy	Particles Range (µm)	
	D	P
9	86.34	124.85
9.5	93.9	136.35
10	101.46	147.85
10.5	109.49	160.145
11	117.52	172.44
11.5	126.02	185.51
12	134.52	198.58
12.5	143.47	212.41
13	152.42	226.24
13.5	161.815	240.789
14	171.21	255.338
14.5	181.04	270.664
15	190.87	285.99

Nuclear cross-sections for ^{192}Os(p,n)^{192}Ir and ^{192}Os(d,2n)^{192}Ir

From TENDL 2017 nuclear cross-section data, the energy value producing the highest cross-section of the reaction can be obtained. This data can be used as a reference to determine the optimum energy for a nuclear reaction although it has to be confirmed with the yield calculation. The nuclear cross-sections of ^{192}Os(p,n)^{192}Ir and ^{192}Os(d,2n)^{192}Ir reactions for proton and deuterons bombardment on ^{192}Os target material can be seen in Fig. 2, where the cross-section increases with increasing energy; however, when it reaches a certain peak value the cross-section shows significant decrease. At the top of this graph the optimum energy that produces the highest cross-section can be determined. For ^{192}Os(p,n)^{192}Ir reaction the optimum energy is between 9 MeV and 11 MeV, with the cross-section values of 59.86 mbarn to 59.23 mbarn. Meanwhile, for ^{192}Os(d,2n)^{192}Ir reaction the optimum energy is between 12 MeV and 14 MeV with cross section values between 586.07 mbarn and 629.11 mbarn.

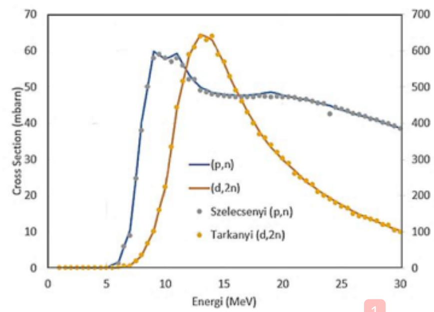

Fig. 2. The comparison of the cross section from. ^{192}Os(p,n)^{192}Ir and ^{192}Os(d,2n)^{192}Ir nuclear reactions.

Radioactivity yields of ^{192}Ir from (p,n) and (d,2n) nuclear reactions

Based on the calculated ^{192}Os(p,n)^{192}Ir and ^{192}Os(d,2n)^{192}Ir yields, as shown in Fig. 3, for proton and deuteron energy ranges between 1 and 30 MeV, the ^{192}Ir yield derived from ^{192}Os(d,2n)^{192}Ir reaction is higher than that of ^{192}Os(p,n)^{192}Ir reaction. At 15 MeV protons and 15 MeV deuterons, the ^{192}Ir yields are 9.73 GBq and 1.86 GBq respectively, whereas for 30 MeV protons and 30 MeV deuterons, the ^{192}Ir yields are 26.14 GBq and 6.79 GBq.

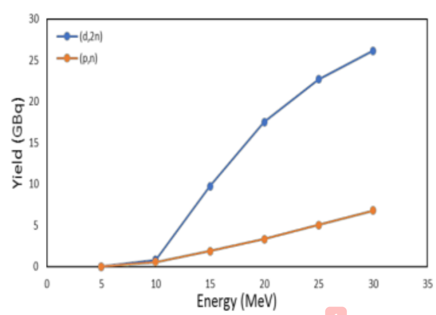

Fig. 3. Calculated ^{192}Ir Yields from ^{192}Os(p,n)^{192}Ir and ^{192}Os(d,2n)^{192}Ir Nuclear Reaction.

As shown in Fig. 4, the calculated production yield in this work has good agreement with the previous works by Higlers and coworkers 2005 [3]. In their work, the production data were based on measurements and assumptions.

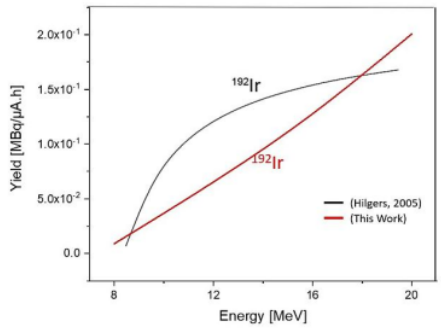

Fig. 4. Calculated ^{192}Ir Yields from ^{192}Os(p,n)^{192}Ir nuclear reaction in this work compare to higlers (2005).

Predicted impurities

In order to predict radionuclide impurities, nuclear cross-sections for various possible protons and deuterons reactions with ^{192}Os were analyzed. As seen in Fig. 5, several nuclear reactions may occur between protons and ^{192}Os target, such as (p,2n), (p,3n), (p,np), (p,d) and (p,α). All of these reactions have significant nuclear cross-sections; thus they could result in radionuclide impurities, such as ^{191m}Ir which is produced from ^{192}Os(p,2n)^{191m}Ir nuclear reaction, ^{190}Ir from ^{192}Os(p,3n)^{190}Ir reaction, ^{191}Os from ^{192}Os(p,np)^{191}Os and ^{192}Os(p,d)^{191}Os reactions, and ^{189}Re from ^{192}Os(p,α)^{189}Re reaction.

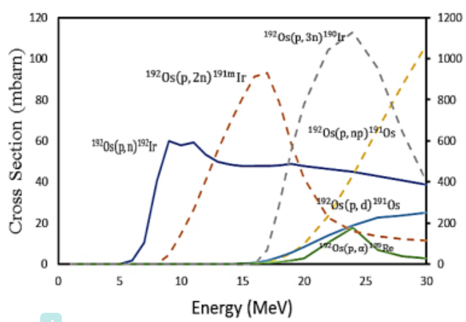

Fig. 5. Nuclear Cross-Sections of (p,2n), (p,3n), (p,np), (p,d) and (p,α) reactions for ^{192}Os Target.

Based on the calculated nuclear cross-sections for (d,2n), (d,3n), (d,p), (d,np) and (d,nd), significant amount of radioactive impurities could be generated when ^{192}Os target is bombarded with deuteron beams. As shown in Fig. 6, the impurities include ^{192}Ir produced by ^{192}Os(d,2n)^{192}Ir reaction, ^{191m}Ir by ^{192}Os(d,3n)^{191m}Ir reaction, ^{193}Os from ^{192}Os(d,p)^{193}Os reaction, ^{192m}Os from ^{192}Os(d,np)^{192m}Os reaction, and ^{191}Os from ^{192}Os(d,nd)^{191}Os reaction.

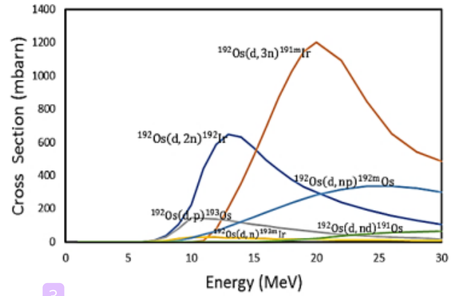

Fig. 6. Nuclear Cross-Sections of (d,2n), (d,3n), (d,np), (d,p), (d,n), and (d,nd) reactions for ^{192}Os Target.

Radionuclide impurities resulted from Os bombardment of protons and deuterons vary from short lived (as short as 4.90 seconds) to long lived (73.83 days) ones. The complete predicted radionuclide impurities are listed in Table 2, which indicates that most of them are β^{-} emitters. In addition, there is no stable isotope predicted to contaminate in the ^{192}Ir production.

Table 2. Various impurities predicted during production of ^{192}Ir radionuclide

Isotope	Nuclear Reaction	Threshold energy (MeV)	Decay mode	Half life
^{191m}Ir	^{192}Os(p,2n)^{191m}Ir	8.0	IT	5.50 s
^{190}Ir	^{192}Os(p,3n)^{190}Ir	16.14	IT	1.12 h
^{191}Os	^{192}Os(p,np)^{191}Os	7.60	β^{-}	15.40 d
^{191}Os	^{192}Os(p,d)^{191}Os	5.36	β^{-}	15.40 d
^{189}Re	^{192}Os(p,α)^{189}Re	0.00	β^{-}	24.30 h
^{192}Ir	^{192}Os(d,2n)^{192}Ir	4.09	β^{-}, EC	73.83 d
^{191m}Ir	^{192}Os(d,3n)^{191m}Ir	10.36	IT	4.90 s
^{193}Os	^{192}Os(d,p)^{193}Os	0.00	β^{-}	29.8 h
^{192m}Os	^{192}Os(d,np)^{192m}Os	2.25	IT	5.9 s
^{191}Os	^{192}Os(d,nd)^{191}Os	7.64	β^{-}	15.4 d

CONCLUSION

Production of ^{192}Ir radionuclide via ^{192}Os(p,n)^{192}Ir and ^{192}Os(d,2n)^{192}Ir nuclear reactions has been theoretically studied using the SRIM 2013 code and TENDL 2017. The SRIM code was used to determine the optimum thickness of ^{192}Os target while TENDL 2017 was employed to calculate the nuclear cross-sections. The radioactivity yields upon the particle bombardment was computed from the SRIM-calculated stopping powers and TENDL nuclear cross-sections. Based on the calculated results, the ^{192}Ir yield derived from ^{192}Os(d,2n)^{192}Ir reaction is higher than that of ^{192}Os(p,n)^{192}Ir reaction. Several radionuclides such as ^{191m}Ir, ^{190}Ir, ^{191}Os and ^{189}Re were predicted to be generated during ^{192}Os(p,n)^{192}Ir reaction for proton incident energy between 1 and 30 MeV, whereas ^{192}Ir, ^{191m}Ir,

^{193}Os, ^{193m}Ir, ^{192m}Os and ^{191}Os radionuclides were expected to contaminate during ^{192}Os(d,2n)^{192}Ir reaction for deuteron energy between 1 and 30 MeV.

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