The implication of Thorium fraction on neutronic parameters of pebble bed reactor

Zuhair*, R. Andika Putra Dwijayanto, Suwoto, Topan Setiadipura Center for Nuclear Reactor Technology and Safety – BATAN Puspiptek Complex, OB No. 80, Serpong, Tangerang Selatan 15310, Indonesia * Corresponding author: zuhair@batan.go.id

Abstract

Thorium abundance in the Earth's crust is estimated to be three to four times higher than uranium. This is one potential advantage of Thorium as a provider of attractive fuel to produce nuclear energy. Fewer transuranics produced by Thorium during the fuel burn up in the reactor may also be another advantage for reducing the long-term burden of high-level long-lived waste. The scope of this paper is to study the implication of Thorium fraction on neutronic parameters of pebble bed reactor. The reactor model of HTR-10 was selected, and the (Th, ²³⁵U)O₂ fuel was used in this study. The MCNP6 code was applied to solve a series of neutron transport calculations with various Thorium fractions in $(Th, 235U)O_2$ fuel based on the ENDF/B-VII library. The calculation results show that the total temperature coefficient of reactivity of Thorium-added pebble bed reactors is generally more negative than those of LEU-fuelled one, except for 10% Thorium fraction. The kinetic parameters, especially prompt neutron lifetime and neutron generation time of pebble bed reactors, are higher, which means the addition of Thorium in the fuel makes the reactor more easily controlled. However, the burn-up calculations show that the introduction of Thorium in the same fuel kernel as LEU within the pebble bed reactor is unable to lengthen the fuel residence time, except for a minimum of 40% Thorium fraction.

Keywords: ENDF/B-VII; MCNP6; neutronic parameter; pebble bed reactor; Thorium.

1. Introduction

Thorium abundance in the Earth's crust is estimated to be three to four times higher than Uranium. It is a potential advantage for Thorium as a provider of attractive fuel to generate nuclear power. Fewer transuranics produced by Thorium during the fuel burn up in the reactor is also another advantage for reducing the long-term burden of long-lived radioactive waste. The material properties of Thorium differ from those of Uranium. Compared to UO₂, Thorium in the chemical form of ThO₂ has favourable thermophysical properties obtained from its lower coefficient of thermal expansion and higher thermal conductivity. ThO₂ has a higher melting point (3370°C) compared with that of UO₂ (2840°C). These beneficial properties make it reasonable to assume that ThO₂-based nuclear fuel has good performance in HTGR

operation with high burnup and very high temperature (GIF, 2010).

Thorium is a naturally occurring radioactive element almost entirely composed of the ²³²Th isotope. Since ²³²Th is fertile, it cannot induce fission directly from thermal neutron capture. Similarly, as Thorium has no naturally occurring fissile isotope, it cannot be enriched the same way Uranium is enriched. Thorium, however, plays an important role as a fertile isotope much like ²³⁸U, if not for better. ²³²Th transmutes into ²³³U, a good nuclear fuel when it captures a neutron. Unlike ²³⁹Pu, which is transmuted from ²³⁸U, ²³³U performs better than ²³⁵U in the thermal spectrum due to higher neutron per fission and lower capture cross-section. Using Thorium in a nuclear reactor requires a fissile isotope to provide neutrons for ²³³U

transmutation. The available fissile options are 235 U, 239 Pu, and 233 U. Introducing these isotopes into Thorium can be done by combining and mixing them with the Thorium to form nuclear fuel in oxide [(Th, 233 U)O₂, (Th, 235 U)O₂, and (Th, 239 Pu) O₂] or carbide [(Th, 233 U)C, (Th, 235 U)C, and (Th, 239 Pu)C] compounds (Björk, 2015; Gintner, 2010).

The scope of this paper is to study the implication of Thorium fraction on neutronic parameters of pebble bed reactor. The reactor model of HTR-10 (Wu et al., 2002) was selected in this study due to the fairly complete data regarding its design, specification, and characteristics, which is important for developing the desired computation model. The (Th,²³⁵U)O₂ fuel was used in this study because its feasibility has been demonstrated in almost all types of nuclear reactors. The two boiling water reactors (BWRs) BORAX IV and Elk River in the USA utilized high density $(Th,^{235}U)O_{2}$ fuel. The pebble bed typed high temperature gas-cooled reactors (HTGRs) have adopted this fuel in German Arbeitsgemeinschaft

Versuchsreaktor (AVR) and Thorium-Hochtemperaturreaktor (THTR). The $(Th,^{235}U)O_2$ fuel has also been employed in the prismatic block typed HTGRs Peach Bottom and Fort St Vrain in the USA. In Canada, the material testing reactors (MTRs) NRU and NRX with pin assemblies have accommodated this fuel for irradiation and testing of few fuel elements (IAEA-TECDOC-1450, 2005).

The Monte Carlo transport code MCNP6 (Goorley et al., 2013) was applied to solve a series of neutron transport calculations with different Thorium fractions in (Th,²³⁵U)O, fuel based on nuclear data library ENDF/B-VII (Chadwick et al., 2006). The neutronic calculated include parameters effective multiplication factor (k_{inf}) , temperature coefficient and of reactivity, kinetic parameters. The concentration of several important isotopes resulting from fuel burnup was estimated using CINDER90 (Wilson et al., 2006) depletion module integrated with MCNP6. The calculation results were summarized to complete an inter-comparison analysis of core physics parameters with different Thorium fractions.

 Table 1. General design characteristic of HTR-10 (Jing & Sun, 1998).

Reactor design	
Reactor nomial power (MWt)	10
Inlet/outlet helium temperature (°C)	250 / 700
Helium pressure (MPa)	3
Helium flow rate (kg/s)	4.3
Number of control rods	10
Number of small absorber balls	7
Number of irradiation experiment	3
Core specification	
Core diameter / height (cm)	180 / 197
The volume of active core (m^3)	5
Fuel to moderator pebbles ratio	57/43
Number of pebbles in full core	27,000
Pebble volume fraction in core	0,61
Average discharged fuel burns up (MWd/tHM)	80,000
Fuel loading mode	Multi-pass

	Fuel	pebble		
The radius of pebble (cm)			3	
The radius of the fuel zon	e (cm)		2.5	
The thickness of the graph	nite shell		0.5	
The density of graphite matrix and shell (g/cm ³)			1.73	
Impurity of natural boron	in a graphite matrix a	ind shell (ppm)	1.3	
	TRISO f	uel particle		
Number of TRISO particles in fuel pebble			8,335	
TRISO packing fraction (%)			5.025	
	Fuel	kernel		
Material			(Th,235U)O2	
Diameter (cm)			0.050	
Density (g/cm ³)			10.41	
Th fraction in fuel (%)			0-50	
Boron impurity in fuel kernel (ppm)			1.3	
Coating layer				
	Material	Thickness (cm)	Density (g/cm ³)	
Buffer	С	0.0090	1.10	
iPyC	С	0.0040	1.90	
SiC	SiC	0.0035	3.18	
oPyC	С	0.0040	1.90	

Tabel 2. General specification of fuel pebble.

2. Materials and methods

2.1 HTR-10 description

HTR-10 is the reference design used in the current study. It is a pebble bed reactor with helium as coolant and graphite as a moderator and a structural material. General design and core characteristics are shown in Tables 1 and 2.

To investigate the effect of the thorium, in the current study, the TRISO particle comprises a spherical fuel kernel of $(Th,^{235}U)O_2$ with a radius of 0.025 cm coated by layers of porous carbon buffer (C), inner pyrolytic carbon (iPyC), silicon carbide (SiC) and outer pyrolytic carbon (oPyC). The thickness data of each layer are 0.009 cm, 0.004 cm, 0.0035 cm, and 0.004 cm, respectively. The buffer layer is functioned to provide a space for localizing and retaining the gaseous fission products. The other three layers are designed to provide a barrier for fission product release even at a very high temperature of 1600 °C. Figure 1 shows the configuration of pebble fuel.

Natural Thorium was used in (Th,²³⁵U)O₂ fuel with fraction varied from 0% to 50%. The fuel with 0% Thorium fraction is treated as the same as current HTR-10 fuel, which is 17%-enriched UO₂ fuel. Various Thorium fractions in (Th,²³⁵U) O₂ fuel is intended to analyze the implications of different fuel compositions on the neutronic parameters of the pebble bed reactor. A study showed that a 30% Thorium fraction for a 15% ²³⁵U enriched fuel was the optimum fraction, and the optimum fraction decreased for lower ²³⁵U enrichment (Irwanto et al., 2012). Besides, this study also showed that a Thorium fraction of more than 50% gave a significant reduction in the burn-up performance of the core. Based on this finding, a maximum of 50% Thorium fraction was set in the current study. An interval of 10% was enough to represent the physical trend of Thorium fraction impact of the neutronic properties of the core.



Fig. 1. Configuration of fuel pebble (Setiadipura et al., 2015).

2.2 Calculation model

The modeling of the pebble bed reactor should consider the double heterogeneity features characterized by the random distribution of both TRISO particles in fuel pebble and the pebbles within the reactor core. MCNP6 offers complex geometry modeling capabilities with high accuracy 3-D calculations of the physical system; thus, it can treat the double heterogeneity problem. MCNP6 is the latest Monte Carlo transport code representing the culmination of many years of efforts to combine all features of MCNP5 and MCNPX codes into one product.

2.2.1 Fuel pebble model

Fuel pebble modeling was preceded by modeling the TRISO particles. Fuel kernel and its coating layers were placed in the center of a simple cubic (SC) lattice with exact dimensions. The SC lattice was chosen because its simplicity and calculation difference with other lattices (BCC, FCC, HCP) are not significant (Wahid *et al.*, 2019). The square pitch of the cubic lattice was calculated to be 0.1988 cm which corresponds to the TRISO packing fraction of 5.0248%. The graphite matrix where TRISO particles are embedded was modeled to occupy a remaining space outside the TRISO particles.

Table 3. Atomic densities of	of (Th, ²³⁵ U)O ₂ fuel kernel	•
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Th fraction	230 _{Th}	232 _{Th}	235 _U	238 _U	16 _O
0	-	-	3.9920×10-3	1.9244×10-2	4.6473×10-2
10	4.7975×10-7	2.3776×10-3	3.5919×10-3	1.7315×10-2	4.6570×10-2
20	9.5923×10-7	4.7538×10-3	3.1919×10-3	1.5387×10-2	4.6667×10-2
30	1.4384×10-6	7.1287×10 ⁻³	2.7921×10 ⁻³	1.3460×10-2	4.6764×10 ⁻²
40	1.9174×10-6	9.5022×10 ⁻³	2.3926×10 ⁻³	1.1534×10-2	4.6861×10-2
50	2.3961×10 ⁻⁶	1.1874×10-2	1.9932×10 ⁻³	9.6087×10 ⁻³	4.6958×10 ⁻²

The model of fuel pebble was constructed by expanding the SC lattice using a repetitive structure so that 8,335 TRISO particles fill the fuel zone of the pebble. The graphite shell was modeled, wrapping the fuel zone to form complete modeling of fuel pebble. Since the packing fraction is small, the TRISO particles in the fuel pebble are relatively distant from each other. The effect of truncated TRISO at the surface between the fuel zone and graphite shell caused by repetitive structure can be ignored. The atomic densities of $(Th,^{235}U)O_2$ fuel kernel with various Thorium fractions are given in Table 3.

2.2.2 Reactor core model

Similar to the fuel pebble model, the reactor core model starts by modeling the pebbles. In this case, BCC lattice was used. Two pebbles in the BCC lattice consist of eight partial moderator pebbles (1/8 pebbles each) at the eight corners and one fuel pebble at the lattice center. The radius of the moderator pebble was adjusted to be 2.731 cm from its actual radius of 3 cm to preserve the fuel to moderator pebbles ratio of 57/43 unchanged. The BCC lattice's square pitch was adjusted to be 6.8772 cm to control the distance between the pebbles defined by a packing fraction of 0.61. The moderator pebbles in the cone area were modeled using BCC lattice also with the same packing fraction of 0.61. The reactor core model containing 27,000 pebbles was constructed by expanding the BCC lattice using a repetitive structure. The truncated pebble at the surface of the reactor core resulted by repetitive structure was eliminated by creating an exclusive zone of 1.71 cm helium encircling the reactor core. The moderator pebbles in the cone area were modeled using the BCC lattice with its actual radius at the center of the lattice and radius of 2.731 cm at the eight corners of the lattice. The combination of three geometry options of LATTICE, FILL, and UNIVERSE provided by MCNP6 was utilized to model fuel pebble and model the reactor core.

The main components of the reactor, such as 20 helium riser columns in the side reflector, 10 control rod columns, 7 absorber ball columns, and 3 experimental columns for irradiation, were modeled using basic surface types of cylinders, planes, and pads according to their geometry and dimensions without assumptions and simplifications. The combination of three geometry options of FILL, UNIVERSE, and TRCL was utilized to model reactor components. The fuel discharge tube and the reflector consisting mostly of boronated carbon block were also modeled in detail. This modeling methodology was developed and already validated using several critical experiments (Lebenhaft, 2001) and has been demonstrated in numerous publications (Alzamly et al., 2020; Zuhair et al., 2020; Zuhair et al., 2019a; Zuhair et al., 2019b; Hosseini & Athari-Allaf, 2016; Hosseini & Athari-Allaf, 2012). The fuel pebble model and reactor core model are illustrated in Figures 2 and 3, respectively.

To solve the criticality problem, in addition to the geometry description and material cards, the KCODE and the KSRC cards are required. These are two main control cards used to specify the MCNP6 criticality source for determining the effective multiplication factor (left) and specify the location of the starting point source for a KCODE calculation, respectively. In the KCODE card, a nominal source size of 10,000 neutron histories, a k_{inf} estimation of 1.0, a skipped initial cycle of 50, and a total running of 250 cycles were simulated. The skipped cycle was used to ensure convergence before starting the average calculation from one cycle to the next cycle. The initial spatial distribution of source points was randomly located inside the fissile materials in the geometry of the reactor. The thermal neutron scattering data $S(\alpha, \beta)$ was applied to take into account the molecular binding affinity in all materials containing graphite which interacts with thermal neutron at the energy region below ~4 eV. The outer boundary condition of the pebble bed reactor was defined to be under vacuum condition. All control rods were set at a full withdrawal position.



Fig. 3. Reactor core model.

3. Results and discussion

The calculation results of the effective multiplication factor (kinf) as the function of the Th fraction are illustrated in Figure 4. For all fractions, the k_{inf} value decreases as the temperature rises. This implies that Thorium addition in the reactor can still maintain a negative temperature coefficient of reactivity (TCR). Furthermore, it was observed that there is a slight increase in negative TCR in most fractions of Thoriumadded fuel. The figure shows that the LEUfueled pebble bed reactor has the highest k_{inf} value compared to Thorium-added ones. The problem is that Thorium-added fuel necessitates the reduction of fissile ²³⁵U, exacerbated by a higher capture crosssection of Th compared to ²³⁸U. Thus, 50% Th fraction understandably has the lowest initial k_{inf}. This happens in most Thoriumfueled reactors, even if the fissile content is kept the same.

Table 4 shows the calculation results of the temperature coefficient of reactivity (TCR) as the function of Thorium fraction. As previously mentioned, a higher Thorium fraction provides more negative TCR. This is especially true for the fuel temperature coefficient (FTC), which is more negative for all Thorium fractions. Meanwhile, moderator temperature coefficient (MTC) is comparatively less negative in thoriumadded pebbled bed reactor up to 30% of Thorium fraction. The MTC is more negative in subsequent fractions. Nevertheless, the less negative MTC is compensated by more negative FTC so that the TCR value of Thorium-added reactors is generally more negative than the LEU-fuelled pebble bed reactor. For exception is at 10% Thorium fraction, which TCR is less negative.

This finding is interesting because, in other studies, Thorium addition is found to be reducing the negative value of TCR (Suwoto et al., 2018; Xia & Li, 2013). Meaning, TCR of Thorium-added pebble bed reactors are less negative than LEU-only reactor. This difference is caused by a reduction in fissile load due to increased Thorium fraction. As the fissile load becomes smaller and the Thorium fraction becomes higher, a smaller fission neutron is absorbed more due to Doppler broadening. However, different TCR characteristics may appear if the fissile load is not altered, i.e., kept the same for all thorium fractions. This possibility must be assessed in future works.



Fig. 4. Effective multiplication factor (k_{eff}) .

Th fraction (%)	Fuel temperature coefficient of reactivity (FTC, %∆k/k)	Moderator temperature coefficient of reactivity (MTC,
		%Δk/k)
0	-1.49759±0.00063	-0.12452±0.00063
10	-1.50838±0.00055	-0.04625±0.00057
20	-1.55688±0.00055	-0.08614±0.00055
30	-1.58196±0.00058	-0.10025±0.00055
40	-1.64961±0.00057	-0.14534±0.00058
50	-1.67050 ± 0.00055	-0.18893±0.00055

Table 4. Temperature coefficient of reactivity.

Table 5. Kinetic parameters.

Th	Effectively delayed	Prompt neutron	Neutron generation
fraction	neutron fraction (β_{eff})	lifetime (ℓ, s)	time (Λ, s)
(%)			
0	0.00668±0.00066	2.2599×10 ⁻³ ±1.6658×10 ⁻⁶	1.34825×10 ⁻³ ±1.351×10 ⁻⁵
10	0.00725 ± 0.00070	2.3504×10-3±1.5504×10-6	1.48911×10 ⁻³ ±1.472×10 ⁻⁵
20	0.00681 ± 0.00064	2.4568×10-3±1.6620×10-6	1.66371×10 ⁻³ ±1.610×10 ⁻⁵
30	0.00645 ± 0.00061	2.5867×10-3±1.7747×10-6	1.85601×10 ⁻³ ±1.736×10 ⁻⁵
40	0.00710 ± 0.00071	2.7432×10-3±1.8522×10-6	2.16895×10 ⁻³ ±2.117×10 ⁻⁵
50	0.00723±0.00075	2.9390×10-3±1.8903×10-6	2.60103×10 ⁻³ ±2.533×10 ⁻⁵



Fig. 5. Infinite multiplication factor (k_{inf}) .

As for kinetic parameters, as shown in Table 5, the calculation results of β_{eff} do not show any particular pattern. Most of the values lie well above β fraction of ²³⁵U except Thorium fraction of 30%. MCNP6 considers uncertainty in kinetic parameters

calculation; thus, the actual value could be higher as well. In general, the β_{eff} in Thorium-added pebble bed reactor is similar to nominal β eff for LEU-fuelled one. Other kinetic parameters show a clearer pattern. The values of prompt neutron lifetime



Fig. 6. Concentration of ²³⁸Pu.



Fig. 7. Concentration of ²³⁹Pu.



Fig. 8. Concentration of ²⁴⁰Pu.



Fig. 9. Concentration of ²⁴¹Pu.





increase with increasing Thorium fraction, which is found in the values of neutron generation time. In this calculation, the temperature was set at 900 K.

Burn-up calculation of infinite multiplication factor (k_{inf}) is shown in Figure 5. As shown in the figure, the LEU-fueled pebble bed reactor has longer effective burn-up days. This is expected since it has the highest fissile load anyway. Increasing Thorium fraction implies that less fissile material can be loaded into the fuel to comply with the 20% enrichment limit. Meanwhile, ²³³U conversion from thorium is insufficient to replace the fissioned ²³⁵U. More on this is explained later.

Another noticeable pattern is apparent after the reactor became subcritical. On the day 2000, 50% Thorium fraction maintains the highest kinf compared to others, while kinf of LEU becomes the lowest. At this point, ²³³U formation from Thorium takes its effect to maintain higher criticality than LEU-fuelled one. However, this is far under critical value. Thus, for this study, the pattern is pointless. In theory, nevertheless, it shows that ²³³U formation took considerably more time than plutonium, which must be put into consideration in further studies.

Plutonium is often considered problematic in the nuclear waste management and proliferation issue. Despite pebble bedtype fuel, a high-temperature reactor is no exception, which is already a strong physical and proliferation barrier. The addition of Thorium is expected to reduce plutonium formation in the spent fuel, thanks to its lower atomic and mass number. Plutonium isotopes evolution for each Thorium fraction is shown in Figures 6 to 10.

For all plutonium isotopes, at day 2000, it is expected that the 0% Thorium fraction has the highest plutonium concentration, despite different buildup patterns. While ²³⁹Pu, ²⁴⁰Pu, and ²⁴¹Pu in the 0% Thorium fraction are constantly the fastest-growing, ²³⁸Pu and ²⁴²Pu are initially building up more slowly. As the Thorium fraction increases, plutonium buildup slows down. Its production peaks at lower burn-up days and then reduces accordingly, apart from ²⁴²Pu has not yet reached its peak.

The aforementioned figures show that Thorium addition is indeed reducing plutonium generation. However, the reduction is not linear to the Thorium fraction in the fuel. The reason is that, even in 50% Thorium fraction, ²³⁹Pu



Fig. 11. Concentration of ²³³U.



Fig. 12. The concentration of ²³³Pa.





into unusable ²³⁴U, reducing ²³³U conversion efficiency. The latter is especially important since the MCNP6 calculation ignores the fuel recirculation system inherent to the pebble bed high-temperature reactor. In reality, fuel pebble is periodically discharged, thus allowing ²³³Pa to decay without capturing neutrons.

The total concentration of ²³³U+²³³Pa is not three times larger than ²³⁹Pu, despite the larger capture cross-section of Thorium. Apart from



Fig. 14. Concentration of ²³⁵U.





the reason mentioned in the previous paragraph, another possible explanation is that Thorium works better in the harder spectrum. In a heavily thermalized reactor like a pebble bed hightemperature reactor, Thorium may not be as effective as capturing a neutron and converting them into fissile material. The possible selfshielding effect might further exacerbate this since the fuel kernel radius is not altered.

Meanwhile, ²³³U and ²³³Pa concentrations peaked approximately at day 800-1400, with the latter peaked at later days. For several Thorium fractions, the peaks are well beyond their criticality limit. Therefore, although the bred ²³³U is considerably large, it is pointless in practical timescale. This shows the constraint of Thorium addition in the same fuel kernel. The 20% enrichment limit necessitates a lower fissile load for a higher Thorium fraction, which is disadvantageous in maintaining long criticality. This factor must be considered when discussing the possibility of Thorium addition in the pebble-bed high-temperature reactor.

Thorium concentration decreased fastest in 50% Thorium fraction, as shown in Figure 13. This is expected since Thorium is the most dominant fertile isotope. Thorium decrease is slower in lower fraction as ²³⁸U became the dxominant fertile.

Figures 14 and 15 illustrate the evolution of ²³⁵U and ²³⁸U. The figures imply that ultimately ²³⁵U is still the main contributor of fission even in Thorium-added hightemperature reactor. ²³⁵U concentration reduction is proportional to k_{inf} change over time. ²³³U conversion from Thorium practically plays almost no part in maintaining criticality, owing to the factors mentioned previously; significantly lower initial fissile load of Thorium-added fuel and longer precursor isotope half-life. In high Thorium fraction (above 30%), the reactor became subcritical long before ²³³U buildup is sufficient to maintain criticality.

Meanwhile, in low Thorium fraction (30% and below), ²³³U buildup is insufficient anyway to compensate for the loss of reactivity due to ²³⁵U depletion. Nonetheless, it is shown that ²³³U and ²³³Pa concentrations are larger than fissile plutonium in 40% and 50% Thorium fraction at all times. The remaining issue is that ²³⁵U loading cannot be higher due to enrichment constraint, prohibiting a high fraction of Thorium from being loaded into the same fuel kernel as LEU.

4. Conclusion

A study on the implication of Thorium fraction on neutronic parameters of pebble bed reactor has been conducted using MCNP6 code based on ENDF/B-VII library. The calculation results show that the addition of Thorium in pebble fuel resulted in a generally more negative total temperature coefficient of reactivity. Among the kinetic parameters, prompt neutron lifetime and neutron generation time are longer, ensuring that the Thorium-added pebble bed reactor can be controlled more easily. Notwithstanding, the advantages of Thorium addition in this study ended here, as burn-up calculations show that Thorium-added fuels are unable to stay critical longer than LEU. This is because fissile loading is reduced when the Thorium fraction increases, whilst ²³³U conversion from Thorium is insufficient to compensate for the lost fissile. To ensure Thorium has a meaningful share in fission events within the fuel, the fraction must be kept at least 40%. If the initial fissile load can be kept the same as LEU-fuelled pebble bed reactor, a minimum of 40% Thorium fraction can potentially lengthen fuel residence time since it contributes more to fission than ²³⁹Pu. Nevertheless, since maintaining the same initial fissile load is constrained by the ²³⁵U enrichment limit, introducing Thorium in the same fuel kernel maybe not the best way to exploit Thorium potential in a pebble bed reactor. Therefore, other strategies must be pursued.

ACKNOWLEDGMENTS

The authors wish to give sincere thanks to Dr. Ir. M. Dhandhang Purwadi, M.T., and Dr. Syaiful Bakhri for their encouragement and support in completing this research work. This work was funded by the Government of the Republic of Indonesia through DIPA PTKRN-BATAN FY 2020.

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Submitted	: 20/06/2020
Revised:	16/09/2020
Accepted:	27/09/2020
DOI:	10.48129/kjs.v48i3.9984