Nuclides Composition of Experimental Power Reactor (RDE) Spent Fuel

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ABSTRACT

The management of spent fuel is an issue of safety for Indonesia in the phase of designing RDE. Several studies regarding spent fuel are limited by geometrical characteristics and number of nuclides library. Therefore, different methodologies utilizing MCNPX2.6.0 were applied to get better information for further research. In this study, a single fuel pebble containing UO2 was burned using 5 cycles of multi-pass loading scheme for 1080 days to obtain the same energy as RDE’s core, which is about 79.90 GWd/MTU. The multiplication factor $k_{inf}$ decreased at each cycle and stopped at 1.14575. Afterwards, the fuel was decayed for 5 years as a representation of cooling time in the spent fuel storage tank. The calculation results in the nuclides composition of the spent fuel after 1080 days of burning and 5 years of cooling containing 241 nuclides, which consist of 21 actinides and 220 nonactinides. Actinides with the highest activity of 8.96 Ci is $^{239}$Pu with mass of 0.0867 g, whose half-life time is 14 years long. Nonactinides with the highest activity of 4.47 Ci is $^{137}$Cs with mass of 0.0514 g, whose half-life time is 30.17 years long. The total activity of spent fuel pebble is 22.9 Ci with total mass of 5.28 g. The mass and activity data of each nuclides contained in the spent pebble will be used in the future research for performing safety analysis of the spent fuel storage tank.

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1. INTRODUCTION

Experimental Power Reactor (Reaktor Daya Eksperimental, RDE) is an Indonesia nuclear reactor project that aim to demonstrate a safe and reliable electricity production [1]. National Nuclear Energy Agency of Indonesia (BATAN) has been carrying out some basic research for designing RDE since 2010. Furthermore, the document of basic engineering design was completed at 2017 while the completion of detail engineering design document is still on progress. The project proposed that hopefully by 2023 the RDE will start its operation [2]. RDE was inspired by the research and development program for the High-Temperature Gas-cooled Reactor (HTGR) to develop HTR-10, the 10 MWth reactor which was launched at Institute of Nuclear and New Energy Technology (INET), Tsinghua University in China about 28 years ago [1, 3-5]. According to the safety demonstration test, HTR-10 had inherent safety features and was recognized as a representative advanced nuclear system for the future [6]. Aside from that, HTR-10 is one of the generation-IV nuclear reactors. On account of this reason, RDE is expected to solve the problem of electricity supply in Indonesia [3].

Inherent safety of nuclear power plant has gotten thoroughly attention after the nuclear accident at Fukushima in Japan. Together with the reactor itself, the spent fuel storage system also has...
to possess inherent safety features [7]. Moreover, the key issues of safety for nuclear reactor, which are reported by the nuclear energy agency (NEA), rely heavily on the management of spent fuel [8]. Generally, spent fuel is considered as a waste but in certain circumstances it is also considered as a potential future energy resource. The management of spent fuel for both definitions must include a spent fuel storage system management [9-10]. The reason behind this management is purely to minimize the risk of radiation by making it isolated to human and the environment for as long as it is still hazardous [11]. The management of spent fuel has also become a safety issue for Indonesia in the phase of designing RDE. Thus, the study of spent fuel storage shall be discussed in several aspects including neutronic aspect. The basic requirement for studying spent fuel storage is the specific information of nuclides contained in the spent fuel. Therefore, an analysis of nuclides composition in spent fuel is essential.

Various studies have been conducted to investigate nuclides in spent fuel [3-4]. They relied merely on ORIGEN2.1 code to characterize the radionuclides of RDE’s spent fuel. The calculation of ORIGEN2.1 can be done in a fast computation time. However, it is limited by one-dimensional of geometrical characteristics along with the numbers of nuclide library that can be handled for about 1,700 nuclides [4, 12-14]. Moreover, in order to get accurate result using ORIGEN2.1, the one-group cross section of the nuclides should be weighted by the neutron flux of the reactor [13-14]. Due to this limitation, CINDER90 code was proposed. CINDER90 can handle three-dimensional of geometry effects and consists of the double amounts of nuclides as in ORIGEN2.1. According to Sung-Min and Myung in 2014, CINDER90 gave the best performance when compared to ORIGEN2.1 [12]. As a coupled code of MCNP with CINDER90, MCNPX2.6.0 has been used in this study to get more reliable and accurate result in characterizing the nuclides of RDE’s spent fuel.

2. THEORY

RDE Fuel Description

The fuel of RDE is exactly the same as the fuel of HTR-10 which is a fuel pebble with TRISO coated particles. Fig. 1 shows the construction of the fuel pebble and the TRISO fuel particles embedded in the fuel zone matrix of the fuel pebble. The kernel of the particle is composed of the fissile material, UO$_2$. A porous carbon buffer zone is designed to provide space for fission-product gases, immediately surrounds the kernel. Moreover, a dense carbon zone, the inner pyrolytic carbon (iPyC) layer, provides a diffusion barrier for the fission products. This is followed by a layer of silicon carbide (SiC), which supplies most of the fuel particle’s strength. Finally, an outer pyrolytic carbon (oPyC) layer protects the SiC layer and provides a further impediment to diffusion [15].

![Fig. 1. The construction of the fuel pebble and the TRISO fuel particles embedded in the fuel zone matrix of the fuel pebble](image)

Nuclear Criticality

A stable fission chain reaction and a constant production rate of fission energy can be sustained by designing a nuclear reactor in such a way so that the rates of neutron absorption and leakage are balanced by the rate of fission neutron production. A given neutron will be born in a fission event and then usually scatter until its eventual death in either an absorption reaction or by leaking out. Certain number of neutrons will be absorbed by fissile or fissionable nuclei and include further fission, hence leading to the birth of new fission neutrons. The ratio of new neutron produced to given neutron is known as multiplication factor $k$ [16]:

$$k = \frac{\text{Number of neutrons in one generation}}{\text{Number of neutron in preceding generation}}$$

(1)

The value of $k$ is categorized into three kinds, which can be seen in Fig. 2.
The equation used in CINDER90 is shown as \[17, 22\]:

\[
\frac{dN_m(t)}{dt} = -N_m(t)\beta_m + \gamma_m + \sum_{k \neq m} N_k(t)\gamma_{k \rightarrow m}
\]

\[
\beta_m = \lambda_m + \phi \sigma^m_u
\]

\[
\gamma_{k \rightarrow m} = \sum_{m \neq k} L_{km}\lambda_k + \sum_{m \neq k} \sum_j \int \gamma_{kmj}(E)\sigma_{k,j}(E)\phi(r, E, t) dE
\]

where \(\beta_m\) is the total transmutation probability of nuclide \(m\), \(\lambda_m\) is the total decay constant of nuclide \(m\), \(\phi\) is the energy integrated neutron flux, \(\sigma^m_u\) is the flux weighted average cross section for neutron absorption by nuclide \(m\), and \(\gamma_{k \rightarrow m}\) is the probability of nuclide \(k\) transmuting by decay or absorption to nuclide \(m\).

The set of differential equations describing all nuclides are coupled because each equation is interrelated and contains atomic density information of other nuclides. This set of coupled differential equations is reduced to a set of independent, linear differential equations using the Markov Chains method [17, 22] shown below:

\[
\frac{dN_i}{dt} = \gamma_i + N_{i-1}(t)\gamma_{i-1} - N_i(t)\beta_i
\]

The solutions of each linear chain determine a partial nuclide density [15, 20] as follows:

\[
N_i(t) = \sum_{m=1}^{n} \prod_{k=m}^{n-1} \gamma_k \left( \frac{1}{\prod_{m=1}^{n} \beta_i} - \sum_{j=m}^{n} \frac{e^{-\beta_j t}}{\prod_{k=m+1}^{n} (\beta_i - \beta_j)} \right)
\]

Partial nuclide densities are then summed to determine the total nuclide density. Each path for each nuclide is defined by the available data.

3. METHODOLOGY

The calculation to predict the nuclides composition of RDE’s spent fuel was carried out by using MCNPX2.6.0 code with ENDF/B-VI nuclear data library. The calculation required a fuel pebble modeling illustrated in Fig. 1. Some studies related to fuel pebble modeling have been done where different kinds of TRISO lattice did not give significant impacts to the burnup performance [21, 23]. In addition to that, a comparison regarding TRISO arrangement in the fuel pebble has been studied where the Auto-filled TRISO in Pebble (ATIP) gave nearly the same result as Organized TRISO in Pebble (OTIP) [5]. Due to this reason, a simple cubic (SC) lattice with ATIP model was used to get an efficient modeling.

The modeling of fuel pebble can be performed through visual editor application presented by MCNP, or by making its own input code using a text editor application. The input code composed of cell card, surface card and data card. The cell card

![Fig. 2. Three kinds of multiplication factor \(k\) based on neutron’s number from time to time [16]](image-url)
holds information about part of the fuel that is bounded by surface described on the surface card as well as the type of material used, material density, cell volume, and the type of particles tracked within those cells. The data card holds specific information about the material used in the previous cell card and type of calculation that user requires MCNP to calculate [21], where in this case is burnup calculation. To get an abundant number of the resulted nuclides in the burnup calculation, Tier 3 parameter was used, given that it comprises plenty of fission products content which have CINDER90 yield information. Apart from that, the threshold of atomic fraction was set from $1.0 \times 10^{-10}$ to $1.0 \times 10^{-20}$ so that more nuclides with smaller atomic fraction will be processed in the burnup calculation.

The burnup scenario used in this research is only for a single fuel pebble. Since the RDE’s core contains 27000 pebbles that will be burned for 1080 days with 10 MW power to obtain energy of 80 GWd/MTU, then a single fuel pebble will be burned for 1080 days with 370 W power to obtain the same energy. Using 5 cycles in the multi-pass loading scheme, a pebble will be burned for 216 days in each cycle. Considering that a pebble will be waiting for its turn in the discharge tube for 40 days [4], then the decay period will also be added at the end of each cycle. Furthermore, another 5 years of decay process will be added as a representation of cooling time in the spent fuel storage tank. The specification of the fuel pebble is explained in Table 1 while the material composition of fuel pebble is defined in Table 2.

**Table 1. Specification of fuel pebble**

| Parameters                                      | Values     |
|------------------------------------------------|------------|
| Density of graphite in matrix and outer shell  | 1.73 g/cm³ |
| Heavy metal (uranium) loading (weight)         | 5.0 g      |
| Uranium enrichment (U-235 / total U by weight) | 17 %       |
| UO₂ density                                    | 10.4 g/cm³ |
| Coating layer materials (starting from kernel) | Buffer/PyC/SiC/PyC |
| Coating layer densities (g/cm³), respectively  | 1.1/1.9/3.18/1.9 |
| Number of TRISO coated particles               | 8335       |
| Thermal energy                                 | 3.64628E-08 MeV |

**Table 2. Material composition of fuel pebble**

| Nuclides | Weight (g) |
|----------|------------|
| $^{235}U$ | 0.85       |
| $^{238}U$ | 4.15       |
| $^{15}O$  | 0.6734     |
| $^{12}C$  | 194.79     |
| $^{28}Si$ | 0.5521     |
| Total    | 200.9345   |

4. **RESULTS AND DISCUSSION**

The modeling of fuel pebble using MCNPX2.6.0 visual editor can be seen in Fig. 3. It appears that the model already satisfied the construction of fuel pebble required for RDE which is illustrated in Fig. 1. Based on the model, the energy result from burnup process is 79.90 GWd/MTU which approximately equal to the energy of RDE’s core. The calculation performed in the fuel pebble is using a reflective boundary condition, hence the multiplication factor value obtained is the $k_{inf}$. The criticality condition of fuel pebble in Fig. 4 shows that $k_{inf}$ decreased at each cycle. This happened because at each burnup process, neutron will be used in a fission event and also will be produced which then usually scatter until it meets its eventual death in either an absorption reaction or by leaking out. Since the boundary of fuel pebble is reflective, there is no neutron leaks out from the system. That’s why even after 1080 days of burning, the multiplication factor value still shows a supercritical condition which is about 1.14575. The $k_{eff}$ value of spent fuel in the storage tank should be in a subcritical condition. Since the value of $k_{inf}$ did not consider the leakage of neutron from the system thus the value of $k_{eff}$ will be much smaller than $k_{inf}$. The value of $k_{eff}$ can be calculated by multiplying the $k_{inf}$ value with the non-leakage probability.
The initial composition of fuel pebble only contains 5.6734 g of UO$_2$ with 0.85 g of $^{235}$U, 4.15 g of $^{238}$U, and 0.6734 g of $^{92}$O. After 1080 days of burning and 5 years of cooling the fuel turns into spent fuel. The spent fuel contains 241 nuclides that could be defined as 21 actinides and 220 nonactinides. Actinides and Nonactinides inventory after 5 years of cooling can be seen in Table 3. Due to the excessive numbers of nonactinides especially the fission products, the data shown in the table is only the nuclides whose activity is higher than 1.0E-10 Ci. All nuclides obtained are basically originated from the fuel itself, UO$_2$, especially the product of fission. Mostly they have medium-to-long half-life because they still existed even after 5 years of cooling.

Table 3. Nuclides composition of spent fuel after 5 years of cooling

| Actinides Inventory | Nuclides     | Mass (g) | Activity (Ci) | Nuclides     | Mass (g) | Activity (Ci) | Nuclides     | Mass (g) | Activity (Ci) |
|---------------------|--------------|----------|---------------|--------------|----------|---------------|--------------|----------|---------------|
| $^{231}$Th         | 5.63E-13     | 2.99E-07 |               | $^{232}$U    | 2.63E-09 | 2.14E-04      | $^{239}$Np   | 3.03E-08 | 7.02E-03      |
| $^{232}$Th         | 2.23E-08     | 2.44E-15 |               | $^{232}$Th   | 2.38E+00 | 8.00E-07      | $^{239}$Pu   | 1.74E-11 | 9.09E-09      |
| $^{233}$U          | 3.46E-11     | 8.00E-07 |               | $^{239}$Pu   | 3.62E-17 | 3.35E-11      | $^{239}$Pu   | 5.04E-03 | 8.63E-02      |
| $^{233}$Pa         | 4.61E-10     | 9.57E-06 |               | $^{239}$Pu   | 1.34E-13 | 1.88E-10      | $^{239}$Pu   | 2.37E-01 | 1.47E-02      |
| $^{234}$U          | 2.24E-04     | 1.39E-06 |               | $^{239}$Pu   | 2.61E-09 | 3.44E-11      | $^{240}$Pu   | 4.01E-02 | 9.10E-03      |
| $^{235}$U          | 1.38E-01     | 2.99E-07 |               | $^{239}$Pu   | 1.36E-02 | 9.57E-06      | $^{241}$Pu   | 8.67E-02 | 8.96E+00      |
| $^{236}$U          | 1.03E-01     | 6.66E-06 |               | $^{239}$Pu   | 8.48E-12 | 2.20E-06      | $^{242}$Pu   | 6.64E-02 | 2.63E-04      |

Nonactinides Inventory

| Nuclides     | Mass (g) | Activity (Ci) | Nuclides     | Mass (g) | Activity (Ci) | Nuclides     | Mass (g) | Activity (Ci) |
|--------------|----------|---------------|--------------|----------|---------------|--------------|----------|---------------|
| $^{14}$C     | 5.33E-10 | 2.37E-09      | $^{103}$Pd   | 1.42E-02 | 7.30E-06      | $^{133}$Ba   | 2.19E-11 | 5.61E-09      |
| $^{35}$Cl    | 1.93E-04 | 2.65E-05      | $^{113}$Ag   | 6.13E-14 | 9.68E-09      | $^{134}$Ba   | 2.84E-13 | 2.08E-08      |
| $^{85}$Kr    | 6.08E-04 | 2.39E-01      | $^{125}$Sn   | 4.73E-10 | 3.89E-06      | $^{134}$La   | 3.74E-14 | 2.08E-08      |
| $^{90}$Mo    | 9.08E-14 | 2.64E-09      | $^{125}$Sn   | 1.25E-03 | 3.54E-05      | $^{135}$Ce   | 6.73E-13 | 1.92E-08      |
| $^{90}$Mo    | 1.67E-02 | 2.35E+00      | $^{125}$Sb   | 1.24E-04 | 1.30E-01      | $^{135}$Ce   | 2.05E-14 | 1.36E-08      |
| $^{90}$Mo    | 4.33E-06 | 2.35E+00      | $^{125}$Sb   | 5.92E-11 | 4.95E-06      | $^{135}$Ce   | 1.29E-04 | 4.12E-01      |
| $^{131}$I    | 4.30E-13 | 1.06E-08      | $^{132}$Te   | 4.55E-14 | 1.38E-08      | $^{135}$Pr   | 2.03E-13 | 1.36E-08      |
| $^{131}$I    | 2.71E-02 | 6.82E-05      | $^{131}$I    | 9.51E-03 | 1.68E-06      | $^{135}$Pr   | 2.90E-15 | 1.05E-08      |
| $^{131}$I    | 3.78E-12 | 8.12E-08      | $^{131}$I    | 8.24E-14 | 1.02E-08      | $^{147}$Nd   | 8.35E-14 | 6.76E-09      |
| $^{131}$I    | 6.25E-08 | 1.17E-08      | $^{131}$I    | 1.37E-15 | 1.42E-08      | $^{147}$Pm   | 1.81E-03 | 1.68E+00      |
| $^{95}$Nb    | 4.38E-12 | 1.72E-07      | $^{133}$Sb   | 1.69E-14 | 1.92E-08      | $^{149}$Pm   | 1.25E-14 | 4.94E-09      |
| $^{95}$Nb    | 3.91E-16 | 1.05E-08      | $^{133}$Sb   | 8.42E-16 | 2.25E-08      | $^{151}$Pm   | 3.25E-15 | 2.38E-09      |
| $^{95}$Mo    | 2.77E-14 | 1.33E-08      | $^{133}$Sb   | 6.26E-15 | 2.21E-08      | $^{151}$Sm   | 3.69E-04 | 9.71E-03      |
| $^{95}$Mo    | 3.17E-02 | 5.43E-04      | $^{133}$Sb   | 1.03E-13 | 1.93E-08      | $^{152}$Eu   | 2.47E-07 | 4.35E-05      |
| $^{109}$Ru   | 7.33E-13 | 2.37E-08      | $^{133}$Sb   | 9.99E-15 | 2.54E-08      | $^{154}$Eu   | 6.61E-04 | 1.79E-01      |
| $^{109}$Ru   | 4.68E-15 | 3.15E-08      | $^{133}$Sb   | 8.18E-04 | 1.06E+00      | $^{155}$Eu   | 1.48E-04 | 7.30E-02      |
| $^{109}$Ru   | 2.51E-04 | 8.32E-01      | $^{133}$Sb   | 1.53E-02 | 1.76E-05      | $^{153}$Cd   | 1.17E-10 | 4.15E-07      |
| $^{105}$Rh   | 3.73E-14 | 3.15E-08      | $^{133}$Sb   | 5.14E-02 | 4.47E+00      | Etc.         | 2.04E+00 | 5.04E-09      |

Total 5.28E+00 2.29E+01
Within all the actinides, $^{241}_{94}Pu$, whose half-life is 14 years, has the highest activity of 8.96 Ci with mass of 0.0867 g. $^{241}_{94}Pu$ is formed when $^{240}_{94}Pu$ captures a neutron, which is yet opposite to $^{240}_{94}Pu$. $^{241}_{94}Pu$ is a fissile material. Among all the nonactinides, $^{137}_{55}Cs$, whose half-life is 30.17 years, has the highest activity of 4.47 Ci with mass of 0.0514 g. $^{90}_{39}Sr$ and $^{90}_{39}Y$ have the same activity of 2.35 Ci which make them the 2nd highest activity after $^{137}_{55}Cs$. Those 3 actinides are the fission products that have vital consequences to human health. Alongside with $^{137}_{55}Cs$, $^{90}_{39}Sr$ becomes a huge concern with its slightly long half-life, which is precisely 28.79 years. Activities history from the top-8 highest nuclides activity after 5 years of cooling is shown in Fig. 5.

![Fig. 5. Top-8 highest activity after 5 years of cooling.](image)

Most of the nuclides shown in Fig. 5 have a fluctuated activity in the burning period and a constant activity at the end of cooling period. The fluctuated activity is occurred due to the additional 40 days of decay processes at the end of each cycle. The zig-zagged curve was formed because the activity went up when the burning process happened and went down when the cooling process happened. The burning period increased the nuclides activity as a result of the production of the nuclides itself while the cooling period decreased the nuclides activity as a result of the decay processes. The increment and decrement mostly caused by the gains and losses in the nuclide density explained in eq. 2. Nuclide with the shortest half-time will has the greatest activity decrement in the decay period. $^{90}_{39}Y$ has the shortest half-time among the other 7 nuclides shown in the graph which is about 64.60 hours. However, since it has a rather small activity then the decrement is also small. The greatest decrement can be seen in $^{144}_{58}Ce$ followed by $^{106}_{44}Ru$ which has the shortest half-life after $^{90}_{39}Y$ which is about 284.91 days and 371.59 days respectively. Furthermore, the constant activity at the end of cooling period happened since the value of losses and gains of nuclides are remained the same.

Within all the nuclides there exist 7 long-lived fission products such as $^{43}_{21}Tc$, $^{126}_{50}Sn$, $^{79}_{37}Se$, with half-life of between 200-300 thousand years, and $^{40}_{20}Ca$, $^{135}_{55}Cs$, $^{107}_{43}Pd$, $^{129}_{53}I$ with half-life of millions of years. Fortunately, those 7 nuclides have a rather small activity. However, some of them, especially $^{43}_{21}Tc$ and $^{129}_{53}I$ have the greatest biohazards as well as neutron capture cross-section. The total activity of spent fuel pebble is 22.9 Ci with total mass of 5.28 g. About 25 nonactinides have an activity below 1.0E-10 Ci and 142 nonactinides have zero activity. Those 167 nuclides are isotopes of Se, Rb, Y, Cd, In, Sn, Sb, Te, I, Cs, La, Ce, Nd, Sm, Eu, Gd, Tb, Dy and etc.

According to Husnayani and Udiyani in 2018, the total activity of RDE spent fuel after 5 years of cooling is 6.43 Ci [3] which is quite small if compared to 22.9 Ci. The comparison of spent fuel activity at the end of burning and cooling period can be seen in Table 4. Overall, the calculation using MCNPX2.6.0 gave a higher activity compared to ORIGEN2.1. This is due to the different methodologies used in the computer code. As explained before, the nuclides discussed in this paper are the nuclides with activity higher than 1.0E-10 while the other one only for important nuclides or nuclides with activity higher than 1.0E-05. Moreover, the nuclear data library used for both studies was different. Based on the ORIGEN user’s manual the nuclear data library used in ORIGEN2.1 is ENDF/B-IV [13-14] while the nuclear data library used in MCNPX2.6.0 is ENDF/B-VI. Therefore, some of nuclides in MCNPX2.6.0 weren’t found in ORIGEN2.1 calculation.

In fact, there is not only one pebble contained in a reactor. There will be thousands of pebbles contained in the reactor core. In relation to the handling process of RDE spent fuel, there will not only contain one spent pebble with 22.9 Ci activity, but there will be thousands of spent pebbles after reactor reach its criticality, which means that the total activity of spent fuel will be multiplied by the number of all spent pebbles.
5. CONCLUSION

The nuclides composition of RDE spent fuel has been analyzed by using MCNPX2.6.0 burnup code. At first the fuel material of UO$_2$, was burned for 1080 days with 370 W power using 5 cycles of multi-pass loading scheme to obtain energy of 79.90 GWd/MTU. After 5 years of cooling, the spent fuel contained 241 nuclides with 21 actinides and 220 nonactinides. Actinides with the highest activity of 8.96 Ci is $^{239}$Pu, with mass of 0.0867 g whose half-life is 14 years. Nonactinides with the highest activity of 4.47 Ci is $^{137}$Cs, with mass of 0.0514 g whose half-life is 30.17 years. The total activity of spent fuel pebble is 22.9 Ci with total mass of 5.28 g. All the mass and activity data from each nuclide obtained from this research can be used in future research for performing safety analysis of the spent fuel storage tank.

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Table 4. Comparison of spent fuel activity using ORIGEN2.1 and MCNPX2.6.0.

| Time (Years) | Activity (Ci) |
|-------------|---------------|
|             | ORIGEN2.1     | MCNPX2.6.0 |
| Burning     |               |
| 1st         | 1.06E+02      | 2.61E+02   |
| 2nd         | 2.19E+01      | 6.03E+01   |
| Cooling     |               |
| 3rd         | 9.34E+00      | 3.10E+01   |
| 4th         | 7.46E+00      | 2.59E+01   |
| 5th         | 6.43E+00      | 2.29E+01   |
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