

ESTIMATION OF THE FISSION PRODUCTS, ACTINIDES AND TRITIUM OF HTR-10

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Given the evolution of High-Temperature Gas-cooled Reactor (HTGR) designs, the source terms for licensing must be developed. There are three potential source terms: fission products, actinides in the fuel and tritium in the coolant. It is necessary to provide first an inventory of the source terms under normal operations. An analysis of source terms has yet to be performed for HTGRs. The previous code, which can estimate the inventory of the source terms for LWRs, cannot be used for HTGRs because the general data of a typical neutron cross-section and flux has not been developed. Thus, this paper uses a combination of the MCNP, ORIGEN, and MONTETEBURNS codes for an estimation of the source terms. A method in which the HTR-10 core is constructed using the unit lattice of a body-centered cubic is developed for core modeling. Based on this modeling method by MCNP, the generation of fission products, actinides and tritium with an increase in the burnup ratio is simulated. The model developed by MCNP appears feasible through a comparison with models developed in previous studies. Continuous fuel management is divided into five periods for the feeding and discharging of fuel pebbles. This discrete fuel management scheme is employed using the MONTEBURNS code. Finally, the work is investigated for 22 isotope fission products of nuclides, 22 actinides in the core, and tritium in the coolant. The activities are mainly distributed within the range of 10^{15} ~ 10^{17} Bq in the equilibrium core of HTR-10. The results appear to be highly probable, and they would be informative when the spent fuel of HTGRs is taken into account. The tritium inventory in the primary coolant is also taken into account without a helium purification system. This article can lay a foundation for future work on analyses of source terms as a platform for safety assessment in HTGRs.

KEYWORDS : Source Term, Fission Product, HTGR

1. INTRODUCTION

The design of high-temperature gas-cooled reactors (HTGRs) has evolved and the safety requirements have been defined [1]; accordingly, the source terms that form the basis of licensing must be developed. Nuclear systems require a reliable inventory of fission products for safety and waste management purposes. There has been an increasing amount of interest in performing burnup-dependent core analyses, including depletion calculations of the inventory of nuclides. The main objectives of depletion calculations are to provide as much detail as possible regarding the composition of irradiated HTGR fuel and to quantify the radioactive inventory for potential source terms.

In order to understand the nature of severe accidents in a HTGR, it is necessary to provide the potential source terms first under normal operations. An analysis of source

terms has yet to be performed for HTGRs. Researchers have studied light water reactors (LWR) [2] extensively and developed various tools of evaluation, such as ORIGEN and CITATION, for those types of reactors. However, these tools are not adoptable, as the typical neutron cross-section data and flux information, together determine the generation of source terms, are not supported in the previous tools. Few arguments have been made for HTGRs, and there is a paucity of experimental measurement data and a lack of simulation results pertaining to unpublished code packages. In particular, a general code of estimation for the source term of HTGRs has not been provided despite the fact that HTGRs are considered to be generation-IV reactors.

Therefore, the purpose of this paper is to calculate a source term inventory of the core by generating information pertaining to the neutron cross-section and flux, particularly on the subject of HTR-10 as a reference reactor, under normal operation.

2. METHOD

2.1 Code Description

Calculations for an inventory estimation of source term are usually performed within the context of the requirements of reactor physics studies, such as in-core fuel management and fuel cycle analysis simulations. Full-core neutronics calculations are strongly dependent on the fuel distribution, the structural materials and the geometry of the core. Due to its importance to accurate calculations in the field of reactor physics, the neutron cross-sections data and flux should be generated according to the core.

Concerning widely used reactors such as LWRs, there are the standard libraries of the Oak Ridge Isotope Generation and depletion (ORIGEN) code which include one-group spectral averaged cross-sections. Spectral-averaged neutron cross-sections are required to solve burnup equations. The spectral-averaged cross-sections depend on the burnup ratio due to changes in the isotope concentrations of the fuel assembly with the burnup. When creating a cross-section library, the spectrum with burnup should be considered. In case of a HTGR, no data library exists; thus, researchers must formulate their own libraries according to the given situation. Table 1 [3] illustrates a simple reason that explains why existing libraries cannot be employed. Within the traditional LWR fuel assembly, the mean free path for both thermal and epithermal neutrons in water is less than 0.8 cm, which is much smaller than the width of the LWR fuel assembly, which is 21.45cm. This indicates that most of the neutrons that originate from one fuel assembly are thermalized locally inside the fuel assembly itself. Thus, the neutron spectrum within the fuel assembly depends on the burnup of the fuel assembly itself. However, the mean free path in graphite is approximately 2.4 cm, which is similar to the fuel sphere radius, 3cm, for pebble bed reactors (PBRs). Hence, the neutrons are very likely to escape from the originating fuel sphere and become thermalized in neighboring fuel spheres. This implies that the neutron cross-section is mostly determined by the burnup of the surrounding

spheres. Therefore, in this study, a neutron cross-section library for PBRs is generated using a Monte Carlo method.

Appropriate computational codes and nuclear data should be used to address this flux generation problem. It may be useful to start by examining the approved codes. Several burnup calculation codes have been developed, and these can be found in the Nuclear Energy Agency data bank. The code combination of MCNP, ORIGEN and MONTEBURNS, which is based on the Monte Carlo method, was chosen for a depletion calculation in the current work, as this code set is more reliable and applicable for HTGRs [4]. The Monte Carlo N Particle transport (MCNP) code is a three-dimensional Monte Carlo transport code that can be used for calculations involving neutron, photon, electron, or coupled neutron/photon/electron transport. It is also capable of calculating the Eigen values for critical systems [5]. The ORIGEN code is a zero-dimension isotope generation and depletion code that is used to calculate the buildup, decay, and processing of radioactive materials [6]. ORIGEN solves the Bateman equation for the concentration N_i of isotope i in a material subject to neutron irradiation. The general differential equation for depletion can be written as follows:

$$\frac{dN_i}{dt} = \sum_{j=1}^N \delta_{ij} \lambda_j N_j + \sum_{k=1}^N f_{ik} N_k \int \varphi(E,t) \sigma_k(E) dE - (\lambda_i + \int \varphi(E,t) \sigma_i(E) dE) N_i \quad (1)$$

Here, λ_j = the decay constant [1/sec],

$\varphi(E,t)$ = the energy-dependent neutron flux [# / cm².sec],

σ_i = the neutron absorption cross-section of the nuclide i [cm²].

MONTEBURNS is a utility code that connects MCNP to ORIGEN and calculates depletion based on the MCNP model. MCNP calculates the one-group microscopic cross-sections and fluxes that are used by ORIGEN in depletion calculations. After performing a depletion calculation, MONTEBURNS extracts the inventory of fission materials from the ORIGEN output file and updates the material compositions automatically in the MCNP input file for

Table 1. The Mean Free Path of the Neutron is Smaller than the Radius of a Pebble Due to a Graphite Moderator

	Quantity	Water	Graphite
Thermal neutron (~0.025eV)	Σ_a (1/m)	2.2	0.029
	Σ_s (1/m)	345.0	41.0
	Mean free path (m)	0.0029	0.024
Epithermal neutron (~0.1eV)	Σ_a (1/m)	2.1	0.029
	Σ_s (1/m)	140.0	41.0
	Mean free path (m)	0.007	0.024

the next burnup step [7]. The workflow by this code set is explained in Fig. 1.

2.2 Core Modeling

There are many HTGRs, but the core of HTR-10 in China, which is a type of PBR, was selected as a reference

benchmark model. IAEA recently published the benchmark problem sets [8], and this sufficient information serves as the guideline to model the HTGR core. The design parameters of HTR-10 are summarized in Table 2. This study models the core through a more accurate approach and deals with some of the core physics benchmark problems proposed for the HTR-10 initial core.

A double-heterogeneous MCNP spherical model was constructed to simulate a core. The first heterogeneity was in a tri-isotropic coated fuel particle (TRISO) forming a fuel pebble and the second heterogeneity was implemented in the lattice of the reactor core. Detailed information of

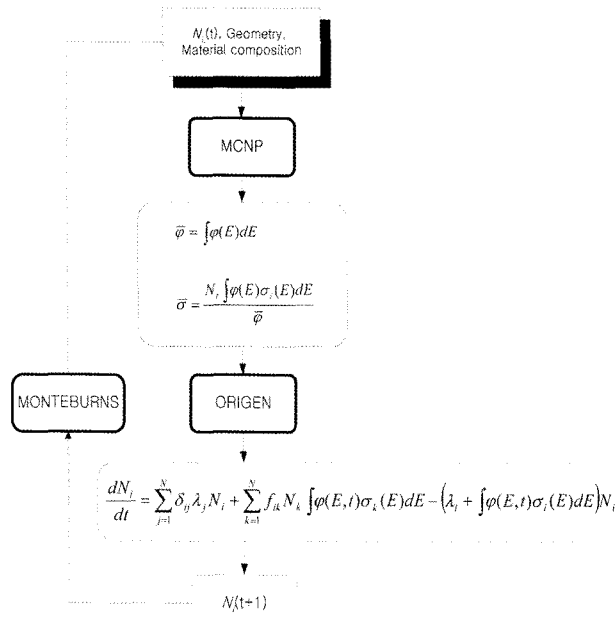


Fig. 1. Workflow of a Burnup Calculation Using with the MCNP-ORIGEN-MONTEBURNS Code

Table 2. Design Parameters of HTR-10

Reactor thermal power [MW]	10
Primary helium pressure	3.0
Reactor core diameter [cm]	180
Core height [cm]	197
Number of control rods in side reflector	10
Number of absorber ball units in side reflector	7
Nuclear fuel	UO ₂
Heavy metal loading per fuel sphere [g]	5
TRISO in Pebble	8335
Enrichment of fresh fuel element [wt%]	17
Number of fuel elements in an equilibrium core	27,000

Table 3. Double Heterogeneity Classification for Core Modeling

	Parameter	Dimension [cm]
Heterogeneity for Pebble	Fuel kernel radius	0.025
	Carbon buffer thickness	0.009
	Inner pyro-carbon thickness	0.004
	Silicon carbide thickness	0.0035
	Outer pyro-carbon thickness	0.004
	TRISO lattice width	0.0976
	No. of TRISO in pebble [#]	8335
	Fuel zone	2.5
Heterogeneity for Core	Pebble graphite shell thickness	0.5
	Filling fraction [unit-less] (reference)	0.65 (~0.61)
	BCC lattice width	6.724
	Fuel : Moderator [unit-less]	57:43
	Radius of a moderator for adjusting the ratio	2.731

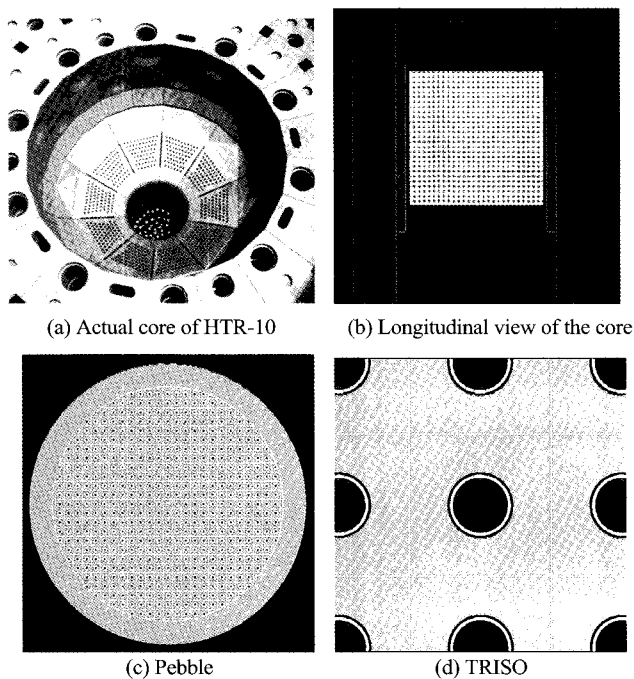


Fig. 2. Double Heterogeneous Core Modeling by MCNP

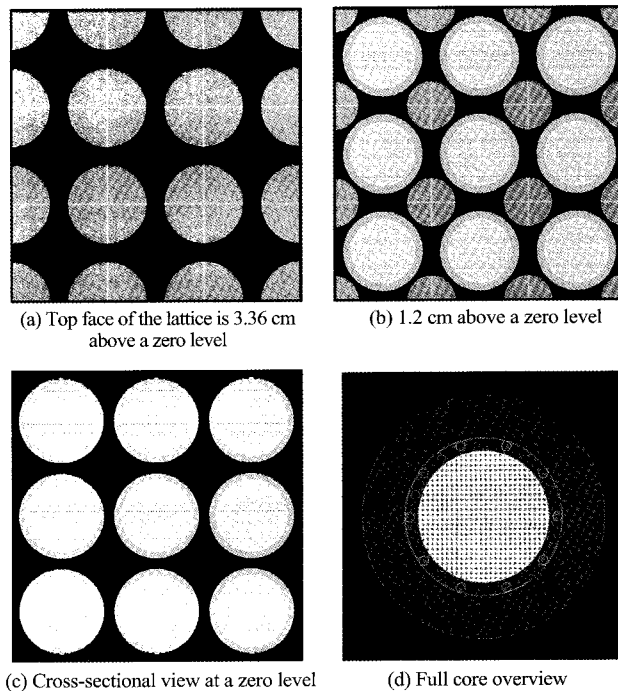


Fig. 3. Four Cross-sectional Images of a BCC Core Lattice. Gray Denotes the Moderator and the Other Areas are the Fuel; (a) Only Moderators, (b) Fuels and Moderators, (c) Only Fuels are Shown

the double heterogeneities is listed in Table 3.

Each TRISO was distributed in the fueled region of the fuel pebbles in the form of a simple-cubic lattice, and the basic unit of the core lattice was constructed in the form of a body-centered cubic (BCC) structure with moderator pebbles of a reduced diameter, which reproduces the specified fuel-to-moderator pebble ratio of 57:43. The double-heterogeneous geometric configurations of the HTR-10 model are shown in Fig. 2, and the typical structure of lattice cells with fuel and a moderator pebble is shown in Fig. 3.

2.3 Fuel Management with an Increase of Burnup

Although shorter burnup intervals can increase the level of accuracy, this benefit requires additional execution time. To balance the tradeoff between accuracy and execution time, it is common to execute MONTEBURNS using, typically, five GWD/MTU burnup steps [4]. This study used a burnup interval of approximately 3.7 GWD/MTU and 23 burnup steps.

The fuel management scheme during operation of up to 80 GWD/ MTU by Yang et al. [9], which is described in Fig. 4, was employed. The continuous fuel management shown in Fig. 4 is divided into the five periods discretely for the MONTEBURNS simulation. The initial 0.43 ratio of the moderator gradually decreases in the core, and only fuel pebbles eventually remain by the fifth stage. It appears reasonable to assume that every fuel pebble should be

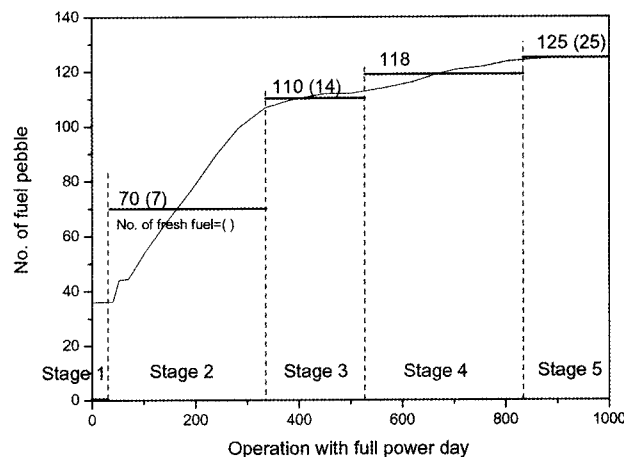


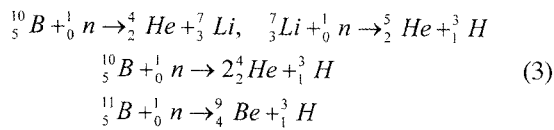
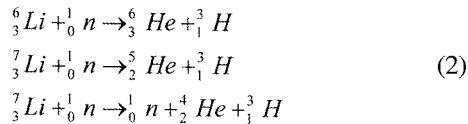
Fig. 4. HTR-10 Fuel Management with Time [9]. The Dotted Lines are the Fuel Supply Intervals

burnt-up at 80 GWD/MTU at the end of the cycle. The PBR design was designed to have a continuous reloading scheme in which unloaded fuel spheres, which have not reached the target burnup amount, are returned to the top of the core.

2.4 Coolant Management for Tritium

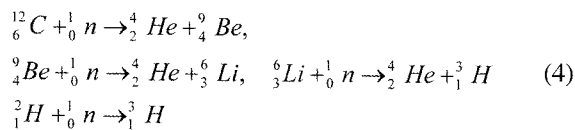
Tritium is considered as the important source term because it can permeate into and through materials, especially metals. This permeation on the heat exchanger causes problems with hydrogen production. Moreover, because tritium behaves chemically as hydrogen, it can undergo isotope exchange reactions with hydrogen-containing chemicals (e.g., H₂O, H₂, or CH₄).

Tritium is produced in reactors by neutron absorption in boron and lithium in the coolant. The reaction is generated by the following mechanisms [10]:



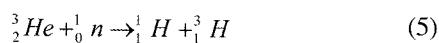
As the concentration of lithium is not known, only boron is considered in equation (3). The reaction of lithium in equation (2) will be assessed in a future study. Boron is not present in coolant but exists in moderators. It was assumed that the dust from the moderators to the coolant was dispersed at a concentration of 0.125ppm [11].

Additionally, tritium generation in the coolant is caused by graphite and hydrogen from coolant impurities of CH₄, H₂O, and H₂:



The anticipated inventories of gaseous impurities in the coolant under normal operation are: H₂O ≤ 0.2 ppm, CO ≤ 3 ppm, N₂ ≤ 1 ppm, H₂ ≤ 3 ppm, CH₄ ≤ 1 ppm [12].

Tritium is also produced in helium coolant, and ³He exists in the amount of 0.000137 % in natural ⁴He.



In equilibrium after the coolant has many passes around the primary circuit, the inventory of tritium must be equal to α ; equation (6) shows this simple process:

$$\alpha = \frac{\sum_{act} \phi_{av} (1 - e^{-\lambda t_i})}{(1 - e^{-\lambda(t_i+t_o)})} \approx \sum_{act} \phi_{av} \frac{t_i}{t_i + t_o} \approx \sum_{act} \phi_{av} \frac{V_{core}}{V_{total}} \quad (6)$$

Here, \sum_{act} = the average macroscopic activation cross-section [cm⁻¹],

ϕ_{av} = the average neutron flux [#/#cm².sec],

λ = the decay constant [/sec]

t_i = the time the coolant spends in the reactor flux [in sec],

t_o = the time the coolant spending in the outer circuit with no flux [sec],

V_{core}/V_{total} = the volume ratio of the core to the total.

The values of \sum_{act} and ϕ_{av} are calculated by MCNP and the depletion calculation is done using ORIGEN. The volume ratio and coolant purification fraction are assumed to be 0.0214 [12] and 1%/day [11], respectively.

3. RESULT

3.1 Validation of MCNP Modeling

The validity of MCNP modeling work can be determined by examining the multiplication factor, k_{eff} , in conjunction with the core height. The method of evaluating whether the modeling works is explained in an IAEA report involving HTR-10 benchmark problems [8], and the question of validation is explained in terms of the estimation of the critical height. A BCC lattice to model the core is used, and the radius of a moderator sphere must be smaller than the original to match the actual fuel-to-moderator ratio, in this case 2.731 cm. Table 4, which compares the results of previous studies and those of the current, confirms that the current modeling provides feasible results. Fig. 5 also shows a comparison of the results of the MCNP simulation with previous studies under the same conditions. The most of the calculation results yield a similar trend. Therefore, these results reflect that the current work can estimate the critical height of the core, showing that the MCNP modeling of the HTR-10 core in the current study is viable.

Table 4. Comparison of the Critical Height [8]

Institution	Method/Code	Critical height [cm]
INET	Experimental	123.06
INET	Diffusion&Transport/V SOP94	125.80
HU	Diffusion&Transport/V SOP94	119.27
NRG	Diffusion & Transport/PANTHER	122.1
FZJ	Monte Carlo/TRIPOLI4	117.37
MIT	Monte Carlo/MCNP4B	127.5
KAIST	Monte Carlo/MCNP	120.95

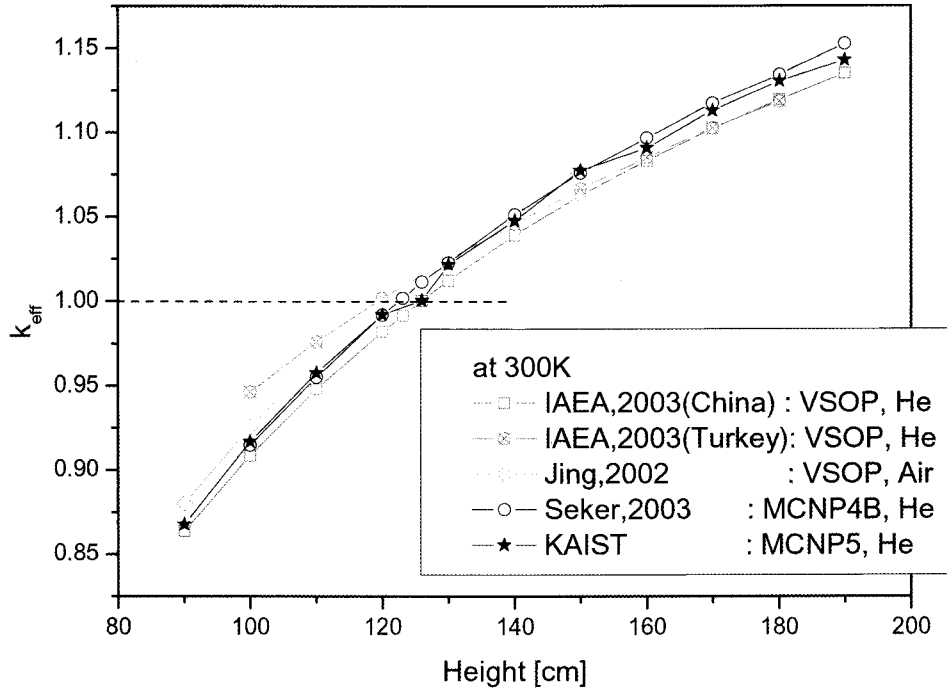


Fig. 5. Variation of k_{eff} with Core Loading [8,13]

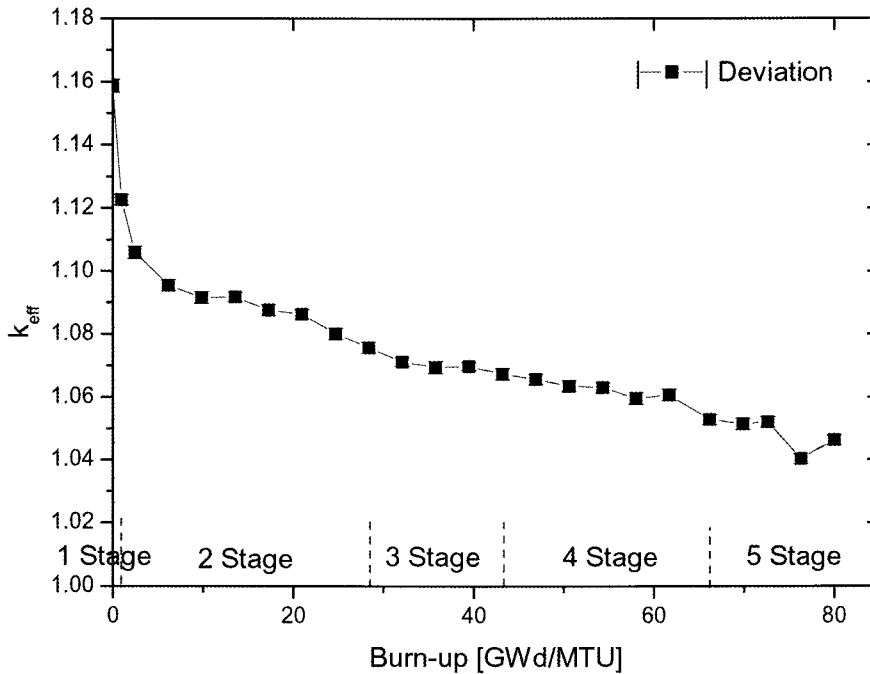


Fig. 6. Reactivity Variation with Time

3.2 Fission Product Inventory

As shown in Fig. 6, the reactor core maintains supercriticality and is operated under full power. As time goes

on with the fuel feed and discharge management described in Fig. 4, k_{eff} gradually declines. The occasional swing under simulated operation may be due to the failure to

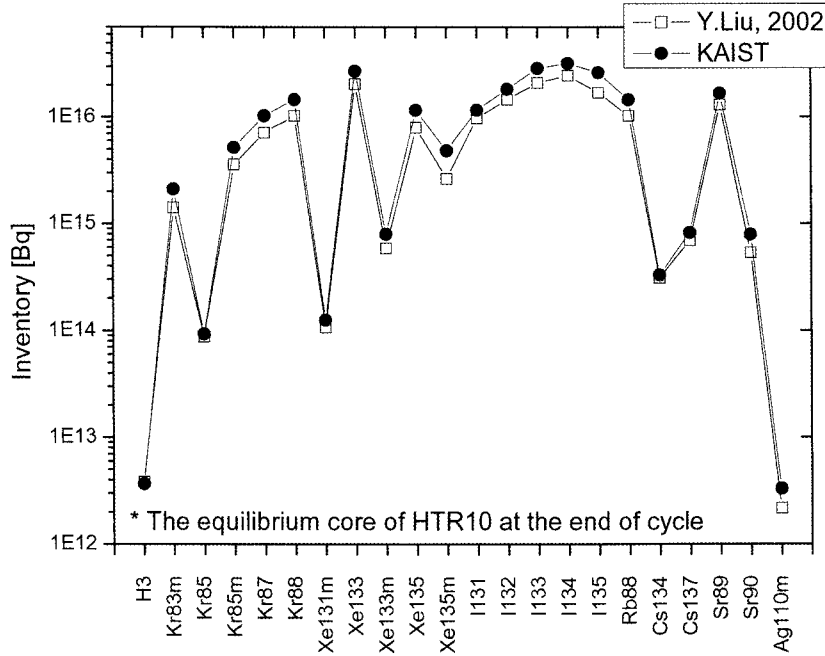


Fig. 7. Comparison of the Inventory of Fission Products [14]

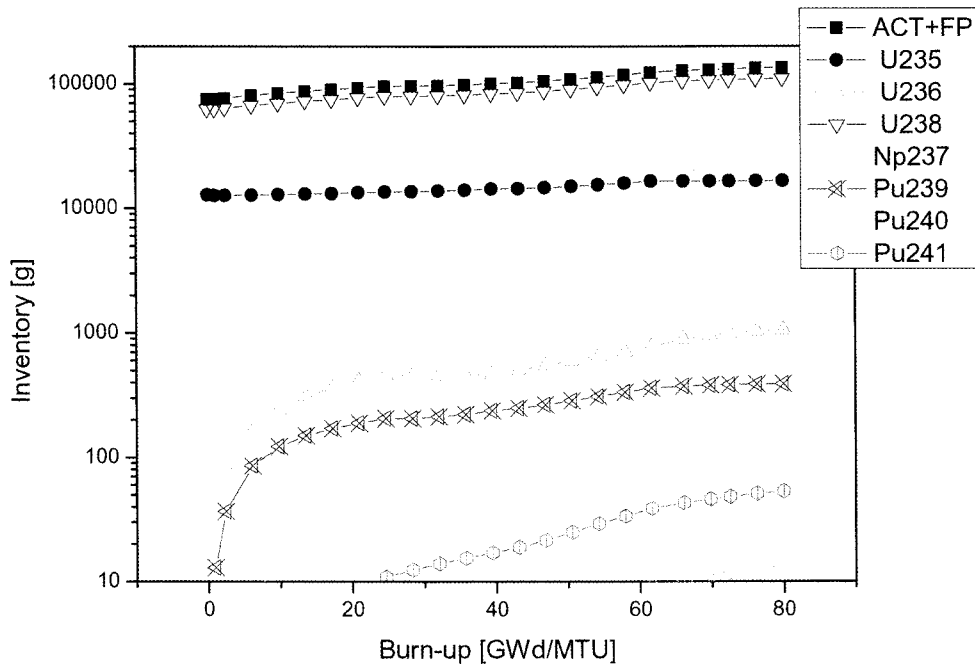


Fig. 8. The Variation of Inventory of Actinide and Fission Products with Time

approach the final convergence value or to a lack of balance in the ratio of the fuel feed and the discharge. It would be interesting to see that the initial peak when the burnup starts is the highest value of k_{eff} . In terms of neutronics, the dramatically steep peak should exist at the beginning,

as an actual reactor is expected to operate smoothly and without problems.

The published inventory data for general HTGRs was difficult to gather. Over ten thousand documents were searched through using keywords for the source term and

fission products. However, only one paper, by Liu and Cao [14], which is insufficient, contained relevant data for HTR-10. The inventories of each isotope in this work and the work by Liu are compared in Fig. 7.

The inventories of 22 nuclide isotopes were compared in relation to the end of the cycle in the equilibrium core of HTR-10. The reference inventory by Liu was calculated solely with ORIGEN2. However, the use of ORIGEN alone can cause erroneous results due to erroneous neutron cross-sections and flux information. Moreover, ORIGEN cannot reflect the variations in the fuel material with time.

Although the trend of the two curves in Fig. 7 appears to be similar, the difference from the two curves comes from Liu's misinterpretation with regard to the unique characteristics of the core itself. In short, this study highlights the need for further investigation due to the paucity of data available for comparison. A considerable amount of research must be conducted in the future.

3.3 Actinide Inventory

It is also possible to confirm from Fig. 8 that the inventories of actinides change as the burnup increases. There is also a rise in the number of actinides and fission products as the core becomes filled with fuel and the moderator is diminished. 22 actinides are traced, but the most of them are small and do not appear in the Figure.

Table 5 compares the actinides produced in the case of HTR-10 and Turkey point after approximately 380 days had passed. Turkey point is a pressurized water reactor (PWR), and the actinides generated in Turkey point can be calculated by ORIGEN-ARP [15]. By increasing the enrichment of ²³⁵U in the case of HTGR, a higher burnup ratio can be achieved and the lower amount of spent material can be generated.

The remarkable point is the amount of isotopes of plutonium, which are perceived as the key issue in nonproliferation issues, in both cases. The most important is ²³⁹Pu as the dominant factor in the determination of the plutonium level. The amount of plutonium in a HTGR is clearly lower than that in a PWR. The ratio of ²³⁹Pu to the total actinides is 0.21% for a HTGR and 0.34% for a PWR, and the ratio of the total amount of plutonium isotopes according to a HTGR over a PWR is only 14%. This reflects the fact that a higher enrichment fuel in a HTGR would be preferred to a lower enrichment fuel in a PWR in terms of proliferation resistance.

Regarding a HTGR, an in-depth study of spent fuel has not been done because the HTGR design of the generation-IV type has not been determined. Therefore, the current work lays the foundation for future spent fuel analysis, providing an inventory of the actinides in fuel pebbles.

3.4 Tritium in Coolant

The variation of tritium inventory with time is shown in Fig. 9. This Figure shows that the activity of tritium in the coolant becomes stable after 400 effective full power

Table 5. Inventory of Actinides in each Equivalent Fuel Region Based on the Reactor Type

Power	10 MW/total	13.5 MW/assembly
Reactor	HTGR	PWR
enrichment	17 %	2.56 %
At time [day]	383	372
GWD/MTU	28.37	9.48
U234 [g]	1.506E-02	8.937E+01
U235	1.476E+04	7.568E+03
U236	4.720E+02	7.751E+02
U237	2.790E-01	4.680E-03
U238	8.527E+04	4.411E+05
U239	4.874E-02	0.000E+00
NP237	4.095E+00	3.629E+01
NP238	7.344E-03	3.967E-09
NP239	6.895E+00	1.810E-06
NP240	8.299E-05	0.000E+00
PU238	2.643E-01	3.654E+00
PU239	2.142E+02	1.555E+03
PU240	5.327E+01	2.995E+02
PU241	1.169E+01	1.264E+02
PU242	1.115E+00	1.315E+01
PU243	8.388E-05	0.000E+00
AM241	1.223E-01	2.318E+00
AM242	3.378E-04	2.452E-07
AM243	2.177E-02	7.067E-01
CM242	1.848E-02	1.207E-01
CM243	4.914E-05	1.026E-03
CM244	1.413E-03	4.791E-02
Total [g]	1.008E+05	4.516E+05

days (EFPD). It then gradually decreases and settles at approximately 8.07×10^9 [Bq/m³] eventually. There are two studies concerning the tritium inventory in the primary coolant of HTR-10 at the end of the cycle; the results of both are compared in Table 6. As mentioned in section 2.4 the main factors affecting tritium generation are the activation cross-section and the neutron flux. The estimation of the neutron flux shown in Fig. 7 is feasible, and the activation cross-sections are constants embedded in ORIGEN2. Therefore, the difference in the results of the current and earlier works may only stem from the conditions rather than the method. In Liu's work [14], helium

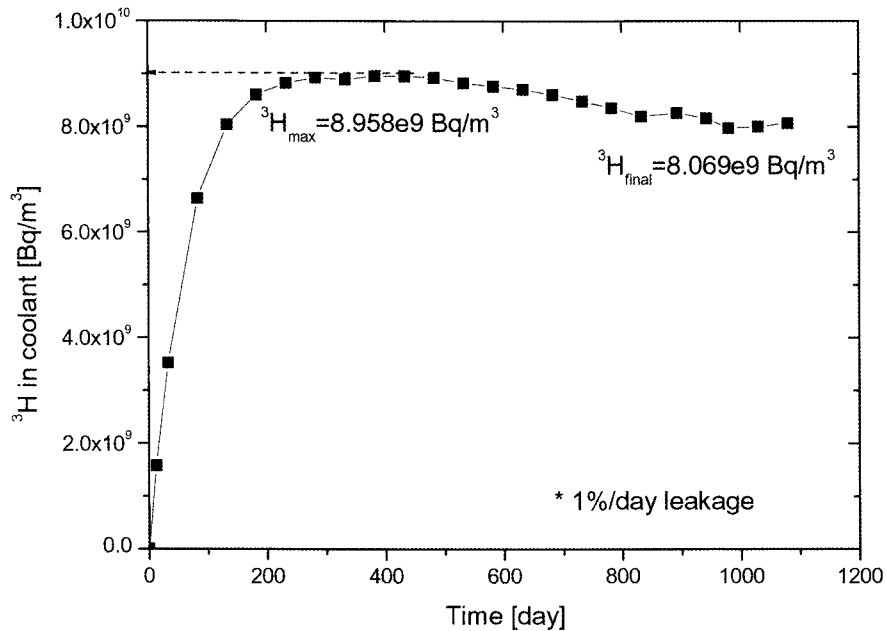


Fig. 9. The Inventory of Tritium Variation with Time

Table 6. Inventory of Tritium in the Primary Coolant at the End of the Cycle

	³ H [Bq/m ³]
KAIST	8.07×10^9
Liu, 2002 [14]	6.10×10^9 *
Liu, 1998 [16]	7.50×10^7

* assumed that the total coolant volume was 93.5 m³

Table 7. The Value of k_{eff} is Dependent on the Temperature in a Helium Atmosphere [8]

T [°C]	INET	FZJ	NRG
20	1.11975	1.12665	1.11759
120	1.11044	1.11331	1.10846
250	1.09596	1.09588	1.09629

purification systems were used. In addition, only tritium by ternary fission and its diffusion were taken into account. This made the inventory lower compared to the result of the current work. There is no description of the inventory by Liu [16]. In addition, the tritium generated in the coolant appears to be overestimated because the helium purification system and permeation are not considered in this study. The effect of introducing a helium purification system will be investigated in a future study.

4. DISCUSSION

It is known that the neutron streaming effect in the coolant channels should be considered in neutronics. It is necessary to quantify the influence of a streaming neutron on the core reactivity, which provides a guideline for the treatment of a streaming effect. However, the neutron streaming effect was estimated to be less than 67 pcm in

a previous study [17]. The neutron streaming effect on reactivity appears to be insignificant and is thus disregarded in the reactor physics analysis for the HTGR core.

In the present work, although considerable effort was made to obtain accurate data, the task of formulating an exact inventory is fraught with difficulties. For example, the reactivity could not be kept constant and no consideration was given to reactivity control or neutron absorber balls. The lack of reactivity control can have an effect on the generation of fission products over time. However, its effect would be small enough to disregard in this study because the fission reaction rate is mainly related to power and not to reactivity. Second, cross-sections for adjusting to the conditions of actual reactors must be provided; these can be generated using the NJOY code for different temperatures with an existing library, such as an evaluated nuclear data file (ENDF). However, generating a cross-section was not discussed here, and the cross-sections used in this study had a default temperature of 300K. The temperature dependence of k_{eff} at under full loading is

listed in Table 7 along with the compensation. Third, there is uncertainty over the feed-discharge rate; the continual process of fuel feeding and discharging is impossible to describe precisely. These four problems need to be explored in the future but are beyond the scope of the present paper. Further studies are needed on different large-scale assessments.

5. CONCLUSION

This article has attempted to sketch out the main method and provide fundamental information related to source terms from which safety assessment would start. It should be stressed that an evaluation of the source term inventory is the first step to be done for nuclear safety.

The main purpose of this work was to explore the source term inventory in a HTGR, particularly a PBR. With help of the combined codes of MCNP, ORIGEN, MONTEBURNS, the author has attempted to obtain the inventory. Regarding the core modeling, the code of MCNP was employed using the BCC lattice, as used in previous studies. Based on the modeling, the generation of fission products with an increase in the burnup amount was simulated. Unfortunately, there are few reference studies available, but the results of the current study seem highly probable. The inventory of actinides in a fuel pebble was also investigated and compared to that in a PWR. The results show a HTGR is more attractive in terms of spent fuel management because the amount of fuel required for the same power is less. It would be informative to take into account an analysis of the spent fuel. Proceeding from this fact, one could logically assume that this inventory simulation is applicable to future spent fuel management schemes, as it provides information pertaining to actinides and fission products. In addition, this paper investigated the inventory of tritium in the coolant. The result implies that coolant purification methods should be improved.

As the actual source-term inventories in HTR-10 are not published, unfortunately, the paucity of references compared to the current work leaves a considerable amount of uncertainty. Although an exact FP inventory could not be obtained due to the uncertainty associated with reactor operation and fuel management, it is hoped that this work will serve as a platform from which an analysis of the source term in HTGR may be undertaken in the future.

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