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ESTIMATION OF GRAPHITE DENSITY AND MECHANICAL STRENGTH OF VHTR DURING AIR-INGRESS ACCIDENT

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ABSTRACT

An air-ingress accident in a VHTR is anticipated to cause severe changes of graphite density and mechanical strength by oxidation process resulting in many side effects. However, quantitative estimations have not been performed yet. In this study, the focus has been on the prediction of graphite density change and mechanical strength using a thermal hydraulic system analysis code. For analysis of the graphite density change, a simple graphite burn-off model was developed based on the similarity concept between parallel electrical circuit and graphite oxidation considering the overall changes of the graphite geometry and density. The developed model was implemented in the VHTR system analysis code, GAMMA, along with other comprehensive graphite oxidation models. GT-MHR 600 MWt reactor was selected as a reference reactor. From the calculation, it was observed that the main oxidation process was derived 5.5 days after the accident following natural convection. The core maximum temperature reached up to 1400 ^oC. However it never exceeded the maximum temperature criteria, 1600 ^oC. According to the calculation results, most of the oxidation occurs in the bottom reflector, so the exothermic heat generated by oxidation did not affect the core heat up. However, the oxidation process highly decreased the density of the bottom reflector making it vulnerable to mechanical stress. In fact, since the bottom reflector sustains the reactor core, the stress is highly concentrated on this part. The calculations were made for up to 11 days after the accident and 4.5% of density decrease was estimated resulting in 25% mechanical strength reduction.

KEYWORDS
VHTR, air-ingress, graphite oxidation, burnoff, mechanical strength

1. INTRODUCTION

The Very High Temperature Gas-Cooled Reactor (VHTR) is one of the promising nuclear reactors due to its passive safety feature and applications to hydrogen production. At this time, the most serious accident considered in this reactor is air-ingress following the large pipe breaks. After the pipe breaks, air in the containment is ingressed into the reactor core and the oxygen gas in the air reacts with the graphite structural materials used for reflector and moderator. A large amount of heat is generated by the exothermic oxidation reaction, increasing the core maximum temperature. From this, the graphite's density is reduced, weakening the structural integrity. In addition, a large amount of undesirable toxic CO gas is generated. Some research has been performed for the air ingress with some preliminary results. According to their results, the maximum core temperature can be maintained below the maximum temperature criteria (1600 ^{o}C) even under the severe chemical reacting conditions ([1], [2], [3]).

In this study, we focused more on the problems of the graphite structural integrity by oxidation, which has not been investigated by other researchers so far. According to Eto and Growcock [4]'s research , the mechanical strength of nuclear graphite is significantly changed by the variation of graphite density, and as a result, approximately 10 percent density change may cause around 50 percent mechanical strength reduction depending on the materials. According to other graphite oxidation research (Fuller and Okho [5], Hino and Hishida [6], Hinsen et al. [7], Katcher and Moorman [8], Kim and NO [9], Moorman [10]), it takes around 7 or 8 days for all the graphite to be transformed into CO or CO_2 gas under 700 C oxidation condition, compared to 2 ~ 3 days over 900 ^{o}C. In reality, the graphite temperature can be increased up to 1500 ^{o}C in the accident conditions, and it means that the mechanical strength of the graphite can be seriously weakened causing the core collapse.

When the core collapses following the graphite oxidation, the fuel particles can directly contact the reactor vessel bottom structure, increasing the temperature up to the melting point of the materials. Then the fractured fuel particles can then exit the reactor vessel. In addition, the fuel on the vessel bottom may react directly with the oxygen in the environment. Therefore, the structural integrity of graphite in the air-ingress accident needs to be investigated.

As mentioned above, the primary factor associated with the graphite mechanical strength is the graphite density. The most essential part in this study is to predict the graphite density variation during the accident. For this, we developed the model to predict the change of graphite density during oxidation and implemented it into the thermal hydraulic system analysis code. Finally, we calculated the variation of graphite density and mechanical strength, which can be used as an indicator to estimate the core structural integrity.

2. GRAPHITE BURN-OFF MODEL

In previous research (Fuller and Okho [5], Mills [11]), the mechanism of the time-dependent graphite oxidation is qualitatively well analyzed and explained. According to their results, the graphite oxidation occurs both on the external surface and in the internal pores. Therefore, the external surface reaction reduces the overall graphite size and shape, and the internal pore reaction changes the graphite density and internal structures. Especially at low temperature (less than 600 ^{o}C), most of the reaction occurs in the internal pores. In this case, at the beginning, the oxidation increases the internal pore size, which increases the active surface area. However, as the oxidation proceeds, the enlarged pores are collapsed and the total active surface area starts to decrease at around 30~40 % burn-off (Fuller et al. [5]), Kim [12]) as shown in Figure 1. At high temperature (more than 1000 ^{o}C), the oxidation reaction simply reduces the size of the graphite, which continuously increases or decreases the active surface area. At the intermediate temperature ranges, both mechanisms occur simultaneously. Even though the graphite oxidation mechanism is well explained, no good model is developed to predict these mechanisms. In this study, we try to accomplish this task.

For modeling, we separated the reaction into the external surface reaction (R_{surr}) and internal pores reaction (R_{bulk}) as follows.

$$R_{bulk} = -\frac{d\rho}{dt}\cdot V \tag{1}$$

$$R_{surr} = -\rho\cdot\frac{dV}{dt} \tag{2}$$

where ρ is graphite density and V is the volume of the graphite materials. Eq. (1) expresses the change of graphite internal density by the internal oxidation, and Eq (2) expresses the change of overall graphite size by the external surface reaction. To solve these equations, we have to first determine the R_{bulk} and R_{surr}. Generally, since the conventional graphite oxidation model calculates the total oxidation rate not separating the internal and external reaction, we need to have additional models to achieve this. To solve this problem, we started from the following global graphite oxidation model suggested by Kim and NO [9].

$$\frac{1}{R_{tot}} = \frac{1}{R_I} + \frac{1}{R_{III}} \tag{3}$$

where R_{tot} is total oxidation rate, and R_I and R_{III} are asymptotic reaction rates at Zone I and III defined as follows.

$$R_I = K_0\cdot\exp(-\frac{E_a}{R\cdot T})\cdot {P_{O2,\infty}}^{0.75}\cdot\theta_0\cdot V\cdot M(B) \tag{4}$$

$$R_{III} = 2\cdot M_c\cdot K_m\cdot(C_{O2,\infty} - C_{O2,0})\cdot A \tag{5}$$

where

E_a : activation energy (kJ/kg-mol)
K_0 : pre-exponential coefficient
R : gas constant (8.314 J/mol K)
T : temperature (K)
P_{O2} : oxygen partial pressure (Pa)

θ_0 : internal surface density (m^2/m^3)
V : volume of graphite (m^3)
$M(B)$: multiplication factor
B : Degree of graphite burnoff ($B = (\rho_{initial} - \rho(t)) / \rho_{initial}$))
M_c : molecular weight of carbon
K_m : mass transfer coefficient of oxygen
$C_{O2,\infty}$: oxygen concentration in the bulk flow (mol/m3)
$C_{O2,0}$: oxygen concentration on the graphite surface (mol/m^3)
A : apparent graphite surface area (m^2)

In Eq. (4), the multiplication factor (M(B)) is the factor for the change of graphite internal structure when the degree of burn-off is B. So, in case where B equals to 0, M(0) becomes unity. This multiplication factor was experimentally determined in this study based on experimental data (Fuller et al. [5], Kim [12]). All data are only for IG-110 graphite material produced by Toyo Tanso. Table I summarizes the physical properties of this graphite. The experimental data are illustrated in Figure 1. In this study, we assumed that the change of internal structure is a function of only degree of burn-off for simplification.

Table I. Properties of IG-110 graphite

Materials	IG-110
Producer	Toyo Tanso (Japan)
Bulk density (g/cm3)	1.75
Young modulus (GPa)	9.6
Compressive strength (MPa)	70.5
Rockwell hardness (MPa)	74.2
Fracture toughness (MPa)	0.82
Thermal conductivity (W/mK)	116
Porosity (vol.%)	21.6
Impurities (ppm)	< 20

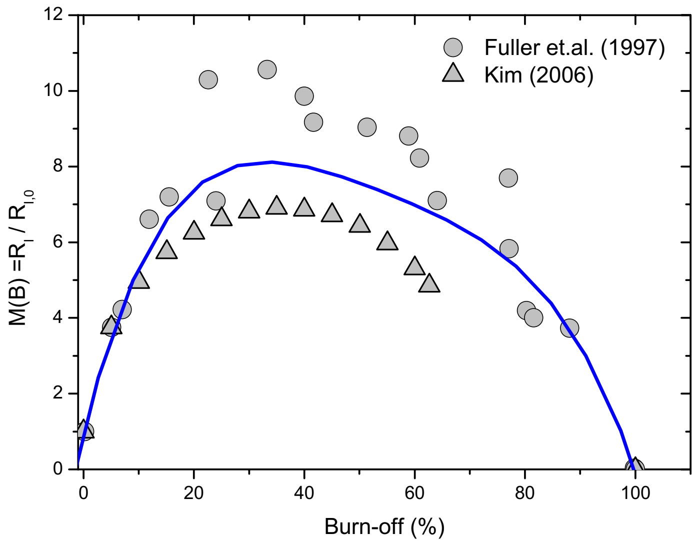

Figure 1 Variation of graphite oxidation rate with time (Fuller et al. [5], Kim (2006)[12]).

To obtain the R_{bulk} and R_{surr}, an analogy study was made between the graphite oxidation model (Eq. (3)) and the electrical circuit shown in Figure 3. The graphite oxidation has similarity with the parallel electrical circuit and the similarity parameters are as summarized in Table II.

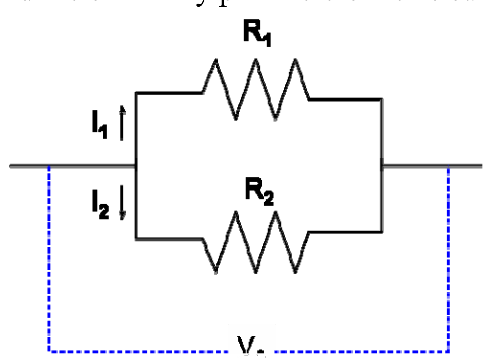

Figure 2 Parallel electrical circuit.

Table II. Similarity parameters between electrical circuit and graphite oxidation.

Electrical Circuit	Graphite Oxidation
V_0	C_{O2}
V_0/R_1, V_0/R_2	R_I, R_{III}
I_1, I_2	R_{surr}, R_{bulk}

According to the analogy, the graphite oxidation rate associated with mass transfer resistance and kinetics can be written as follows, respectively.

$$R_{mass\ resist} = F \cdot R_{tot} \tag{6}$$

$$R_{kinetic\ resist} = (1 - F) \cdot R_{tot} \tag{7}$$

where,

$$F = \frac{1}{R_{III}} / \left(\frac{1}{R_I} + \frac{1}{R_{III}} \right) \tag{8}$$

In this study, we assumed that the reaction by the mass transfer resistance occurs on the graphite surface and the reaction by the kinetics occurs in the internal pores following the similarity between current and asymptotic reaction rates in Table I. Based on the similarity, this assumption is quite reasonable. Therefore, the external surface reaction and internal pore reaction can be re-written as follows.

$$R_{surr} = F \cdot R_{tot} \tag{9}$$

$$R_{bulk} = (1 - F) \cdot R_{tot} \tag{10}$$

3. AIR-INGRESS ANALYSIS AND BURNOFF CALCULATION OF REFERENCE VHTR

In this study, we implemented the newly developed graphite burn-off model into the VHTR system analysis code, GAMMA, developed by KAIST (Korea Advanced Institute of Science and Technology) and INL (Idaho National Laboratory) for analyzing VHTR air-ingress accidents. GT-MHR 600 MWt reactor was selected as a reference reactor. Figure 3 shows the schematics of GT-MHR 600 MWt. In GT-MHR 600 MWt, the coolant enters the core at 490 ^{o}C with 320 kg/s and flows out at 850 ^{o}C. 102 hexagonal fuel elements were built in this model and the reflector graphite block is located at the outside of the active core. Air cooling RCCS passively removes the decay heat. Figure 4 shows the schematic of GAMMA modeling for GT-MHR 600 MWt. To get a good prediction for graphite burn-off, the graphite oxidation rate should be predicted accurately. Table III summarizes the graphite oxidation models used in this calculation. These oxidation models are well validated by comparisons with the experimental data implemented into GAMMA code in the previous research (Lim and NO [13]).

Table III. Graphite oxidation models in GAMMA code.

Total Reaction Rate	$\frac{1}{R_{tot}} = \frac{1}{R_I} + \frac{1}{R_{III}}$
Kinetics model (R_I) (Zone I)	$R_I = 2552000 \cdot \exp(-\frac{218000}{R \cdot T}) \cdot P_{O2,\infty}{}^{0.75} \cdot V \cdot M(B)$ For IG-110 graphite (Kim et al. (2006))
Mass transfer model (R_{III}) (Zone III)	$R_{III} = 2 \cdot M_c \cdot K_m \cdot (C_{O2,\infty} - C_{O2,0}) \cdot A$ where $\frac{h}{\rho v_\infty C_p} (\mathrm{Pr})^{2/3} = \frac{K_m}{v_\infty} (Sc)^{2/3}$ (heat/mass transfer analogy)
CO/CO$_2$ ratio	$f_{CO/CO2} = 7396 \cdot \exp(-\frac{69604}{R \cdot T})$ For IG-110 graphite (Kim and NO (2006))
Heat Release	$\dot{Q} = \sum_x \Delta h_{fs} R_{total}$

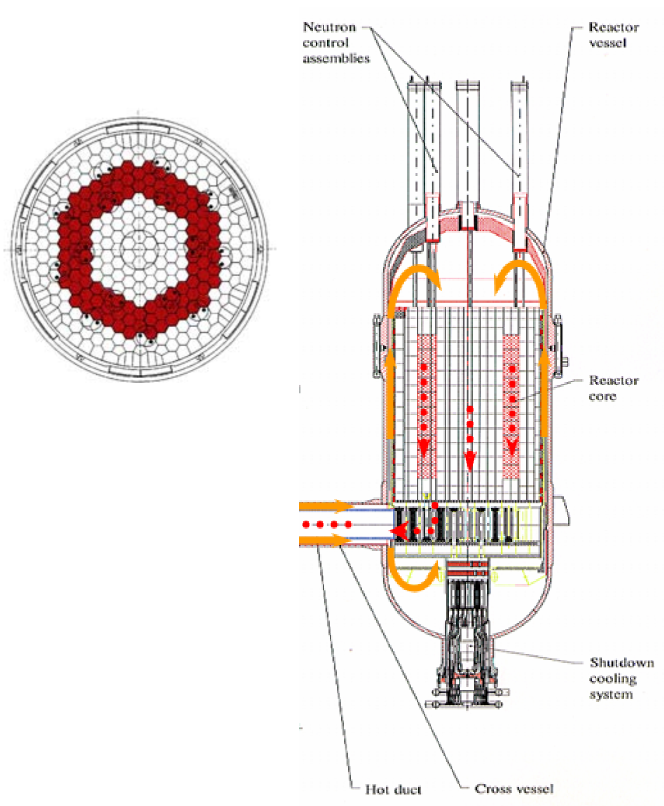

Figure 3 Schematics of GT-MHR 600 MWt.

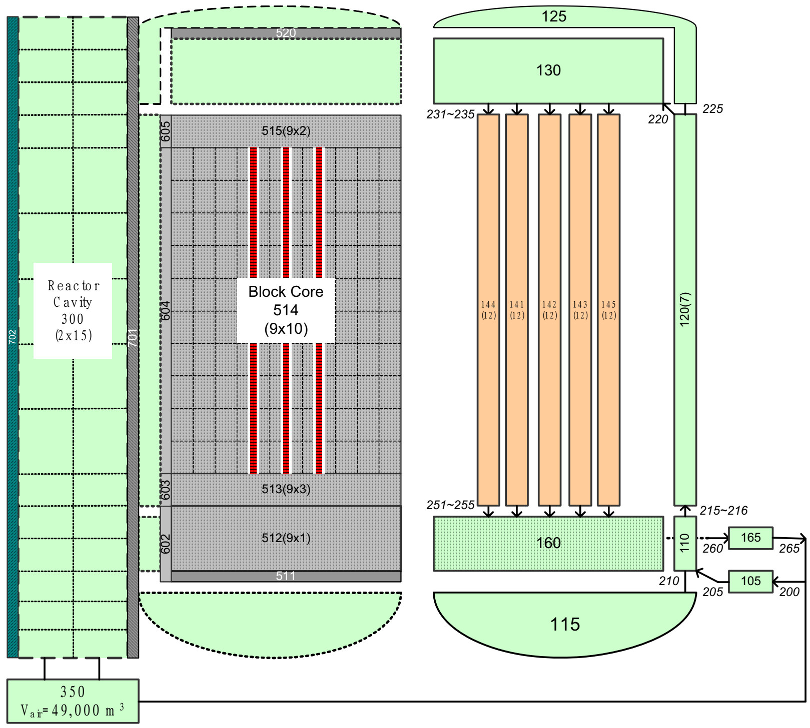

Figure 4 GT-MHR 600 MWt GAMMA Nodalization.

The calculated results are summarized as follows. Figure 5 shows the transient flow rate at the broken pipe. As shown in this figure, a sudden change of flow rate occurs at around 150 hr following the pipe break. It is the important signal for the onset of natural circulation. Figure 6 shows the variation of temperatures in several locations of the reactor core. The maximum temperature was observed around the center of the reactor core approximately 70 hrs after the loss-of-coolant-accident (LOCA). After the peaking point, the temperature continuously decreased. The core maximum temperature reached up to 1430 ^{o}C, which is lower than the maximum temperature criteria of 1600 ^{o}C. At around 150 hrs, a sudden change of temperature in the reflector was observed. This is due to the heat generated by graphite oxidation following the air-ingress with the natural convection. At the core, however, the effects of natural convection and graphite oxidation were negligible because the most graphite oxidation occurs at the bottom reflector. From this result, we expect that the temperature increase by graphite oxidation at the core is negligible and the maximum temperature will not exceed the temperature criteria.

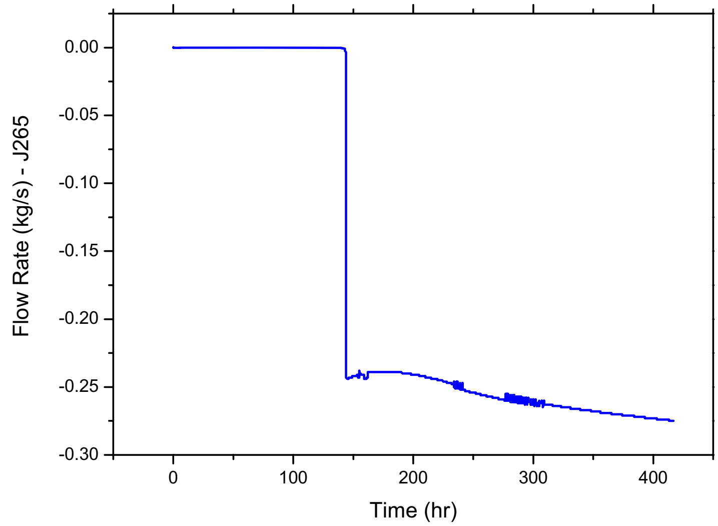

Figure 5 GT-MHR 600 MWt Air-ingress analysis (Flow Rate)

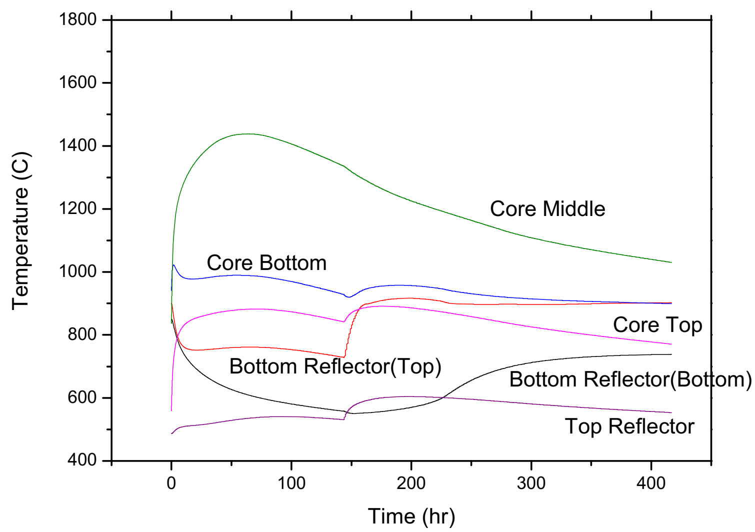

Figure 6 GT-MHR 600 MWt Air-ingress analysis (Temperature).

As indicated in the preceding section, the main purpose of this study was to predict the variation of graphite density and results are shown in Figure 7. As shown in this figure, the significant change in graphite density occurred at 150 hrs after the accident with natural convection. As expected from the temperature results, the oxidation reaction didn't occur at the core and top reflector. At the bottom reflector however, the oxidation severely changed the graphite density. Our calculation continued until 450 hrs (17.5 days), and as a result, the maximum density decrease was predicted up to 5.3 %.

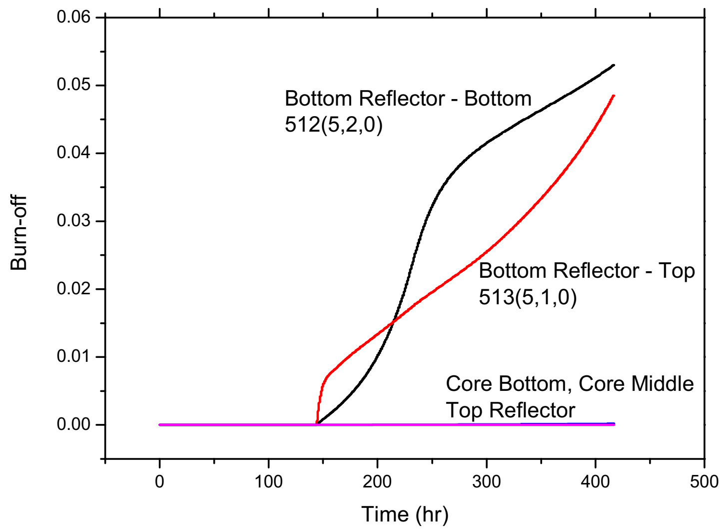

Figure 7 GT-MHR 600 MWt Air-ingress analysis (Burn-off (graphite density)).

From this result, the change of mechanical strength was calculated by the following equation suggested by Eto and Growcock (1981).

$$\frac{S}{S_0} = \left(\frac{\rho}{\rho_0}\right)^{6.08} \text{(for IG-110)} \quad (11)$$

Figure 8 shows the result. According to this calculation, maximum decrease of mechanical strength was 25 %.

Figure 9 shows the change of void fraction in graphite structure with time. The increase of void fraction means that the flow channel is enlarged by the oxidation on the surface. The size of channel is increased and the total volume of graphite decreases with time (refer to Figure 10). It makes the graphite structure very weak and concentrates the stress on a small area. At the core and top reflector, since there is little oxidation reaction, the change of void fraction is not observed. However, in the bottom reflector, significant change of void fraction was predicted. At the top of the bottom reflector, the void fraction is increased from 0.2 to 0.75 after 400 hrs. It means only 30 % of graphite remained at this time shaping like a skeleton. In addition, if the effect of 25 % mechanical strength reduction is added here, the structural integrity of reactor core is questionable. The reason why the oxidation reaction is concentrated on the top of the bottom reflector is that the temperature at this location is increased up to 900 ^{o}C and consumes most of oxygen molecules ingressed. Due to the high temperature, the external surface reaction is preferred here.

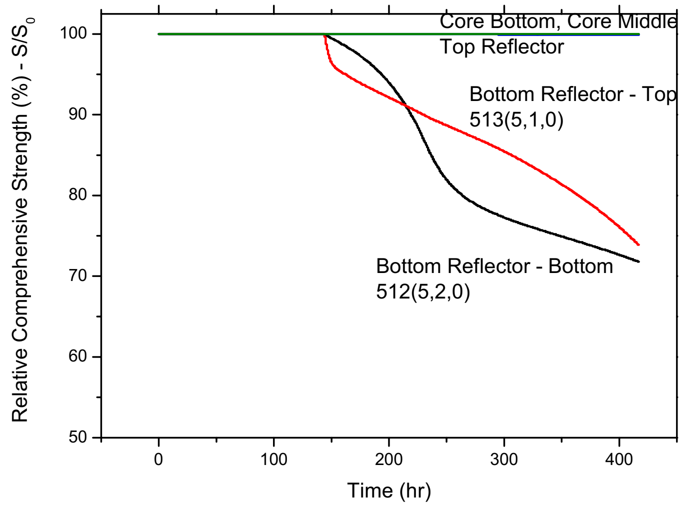

Figure 8 GT-MHR 600 MWt Air-ingress analysis (Mechanical Strength).

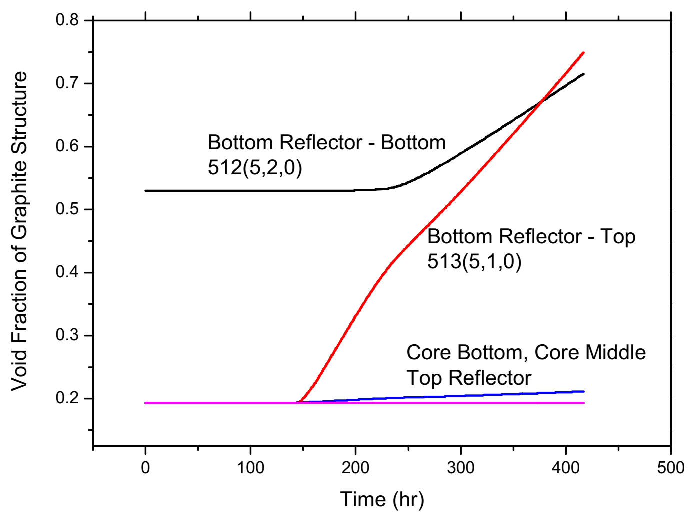

Figure 9 GT-MHR 600 MWt Air-ingress analysis (Void fraction of graphite structure).

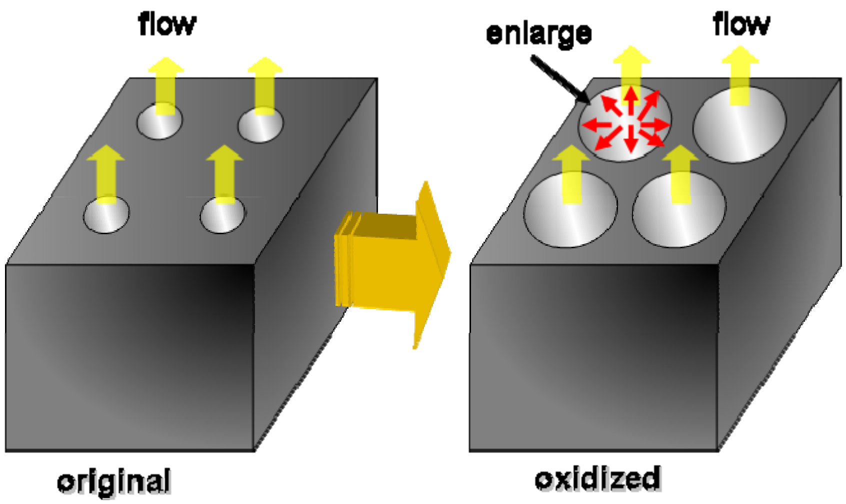

Figure 10. Change of graphite structure void fraction by oxidation.

4. CONCLUSIONS

An air-ingress accident in a VHTR is anticipated to cause severe changes of graphite density and mechanical strength by oxidation process resulting in many side effects. However, quantitative estimation have not been performed yet. In this study, the focus has been on the prediction of graphite density change and mechanical strength using a thermal hydraulic system analysis code.

For analysis of the graphite density change, a simple graphite burn-off model was developed based on the analogy concept between a parallel electrical circuit and graphite oxidation considering the overall changes of the graphite geometry and density. The developed model was implemented in the VHTR system analysis code, GAMMA, along with other comprehensive graphite oxidation models. GT-MHR 600 MWt reactor was selected as a reference reactor. From the calculation, it was observed that the main oxidation process occurs 5.5 days following the LOCA. The core maximum temperature reached up to 1400 ^{o}C. However it never exceeded the maximum temperature criteria of 1600 ^{o}C. According to the calculation results, most oxidation occurs in the bottom reflector, so the exothermic heat generated by oxidation did not affect the core heat up significantly. However, the oxidation process highly decreased the density of the bottom reflector, making it vulnerable to mechanical stress failure. In fact, since the bottom reflector sustains the reactor core, the stress is highly concentrated on this part. The calculation continued until 11 days after the accident, and finally 4.5% density decrease was estimated, resulting in 30% mechanical strength reduction. One of the most important results is the reduction of 75% graphite volume, which might cause serious structural problems. The data and results predicted here can be used for the estimation of graphite structural integrity later, and it is highly recommended.

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