論 文 の 内 容 の 要 旨

論文題目

Assessment of graphite oxidization model for HTGR air ingress accident

(高温ガス炉空気侵入事故時の黒鉛酸化の評価)

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The development of high temperature gas-cooled reactor (HTGR) was started from 1950s, and recently, the technology has attracted renew interest because of its inherent safety features and its potential of cogenerating of electricity and high temperature process heat to produce hydrogen. Two HTGR test reactors are under active research and development. One is the 30MWth High Temperature engineering Test Reactor (HTTR) in Japan, the other is the 10MWth High Temperature Reactor in China (HTR-10).

The key feature of HTGR is that the reactor is operated at high temperature compared to other reactor designs. The major characteristic of HTGR technology is the helium coolant, graphite moderator and the multi-coated fuel particle design. The helium coolant is chemical inert to core components and remains single phase under all conditions. The graphite has high strength, stability, heat capacity and thermal conductivity under elevated temperature. The fuel deign, which is the refractory coated fuel particles dispersed in a graphite matrix, allows retaining fission products up to 1600˚C. A unique feature of HTGR is that the reactor core has low power density, which is effective for rejecting decay heat passively. Specifically, the decay heat can be passively transferred by natural means (conduction, natural convection and thermal radiation) to reactor pressure vessel and then pass to the environment through thermal radiation.

At present, the most critical accident considered for HTGR design is an air ingress, which caused by

the guillotine type break of main coolant pipe. It starts with helium depressurization, after which air is anticipated to enter the core through the break leading to oxidation of in-core graphite structures and fuel. Such a situation would have serious consequences including temperature increase due to exothermic oxidation reaction, mechanical degradation of graphite structures and accumulation of explosive CO gas in the reactor.

To understand the consequence of air-ingress, the mechanisms of graphite oxidation need to be studied first. Extensive studies regarding this topic were carried out for decades, and following conclusion were drawn. The reaction can be classified into three regime based on temperature, which are the chemical kinetic controlled regime, the in-pore diffusion controlled regime and the mass transfer controlled regime. Also, it is found that in chemical kinetic controlled regime, the oxidation rate strongly dependent on the graphite burn-off with the maximum value found at 30%~40%.

Given the potential hazards and risks caused by air ingress, it is critical to improve the HTGR safety under such event of accident, which serves as the ultimate purpose of present study. The safety of HTGR in the event of air ingress can be improved from different aspects, for instance, developing oxidation resistance core graphites, exploring accident tolerant fuel designs, proposing countermeasures that limit the amount of air ingress into the reactor code, etc. Nevertheless, the basic principle of any studies with respect to HTGR air ingress safety evaluation would be performing HTGR safety analysis so that core behaviors, especially the transient of fuel temperature, can be predicted and analyzed during the accident. To that end, the primary objective of this study was to improve the safety analysis of HTGR in an event of air ingress. To accomplish this goal, the present study was carried out in the following three aspects:

1) Validation experiment: Validation experiment plays a key role in quantitative accuracy assessment of safety analysis codes. In present study, a transient graphite oxidation experiment was conducted with grade IG-110 graphite. The experiment is designed to induce the major phenomena occurring during an event of air ingress, which includes the oxidation process of graphite accompanied by multiple modes of heat transfer such as thermal radiation and convective cooling. During the experiment, the gas mixtures of air and N2 were injected from the bottom of the test section after a steady state temperature condition was attained. Graphite temperate (950˚C, 1150˚C) and oxygen mole fraction (5%~21%) were selected as the main experiential variables.
2) IG-110 emissivity measurement: To investigate how oxidation could affect the radiation heat transfer among graphite components, the normal spectral (λ=1.65μm) emissivity of non-oxidized and oxidized IG-110 specimen was measured in a temperature range of 500˚C to 1000˚C utilizing an inferred thermometer and a K-type thermocouple. The measurements

provide the HTGR safety analysis codes with the emissivities of oxidized and non-oxidized IG-110 graphite in a wide temperature range, which enables accurate prediction of thermal radiation heat transfer among graphite components during air ingress.

3) REALP5/SCDAP code modification: RELAP5/SCDAP code was improved for analysis of HTGR. Specifically, the thermodynamic and transport properties of O_2, CO_2 and CO were added to RELAP5/SCDAP non-condensable database to support the analysis of air ingress. Major chemical reactions and their rate equations were finalized after extensive literature review. A chemical equilibrium graphite oxidation model (of IG-110) and a CO combustion model were incorporated into the code. For the graphite oxidation model, the multiplication factor M(B), which is a correlation between burn-off and reaction rate, was modified based on six individual graphite oxidation experiments. The code was validated against three experiments, which were conducted by Chi et al.[1], Choi et al.[2] and Ogawa et al.[3], that cover the entire temperature range of HTGR. An assessment of the code performance (i.e. with the correct trends and magnitudes) with respect to the transient analysis of graphite oxidation in conjunction with multiple modes of heat transfer was carried out by comparing the simulation with the validation experiment conducted in present study.

The conclusions of each part are summarized as follow:
Validation experiment: A transient graphite oxidation experiment was successfully designed and carried out. In the experiment, two major phenomena occurring during an event of air ingress, which are the process of graphite oxidation and radiation heat transfer, were induced and adequately captured. Two forms of data, namely the transient temperature and the weight loss of IG-110 graphite specimen were measured to support the retrospective analysis of the experiment. Moreover, a transient graphite oxidant database was established based on the seven cases conducted at the high temperature regime (950˚C and 1150˚C) over a wide range of oxygen concentration (5% ~21%). The experimental data can be used for validation of numerical modeling of transient graphite oxidation process in conjunction with multimode heat transfers and quantitative evaluation of HTGR safety analysis code in transient graphite oxidation analysis.

IG-110 emissivity measurement: The normal special emissivity (λ=1.65μm) of IG-110 graphite was measured over a temperature range of 500˚C to 1000˚C. The measurement was performed for both non-oxidized and oxidized rectangular bar specimen. The experimental results lead to the following conclusions:

- Oxidized IG-110 graphite is of rougher surface and higher emissivity. This implies that during an event of air ingress, the graphite core components oxidized by the ingress air are of higher emissivity, which means that they can be more efficiently cooled down by radiation heat transfer.

- IG-110 graphite has weak negative temperature dependency of emissivity.
- It has been made clear that the commonly used IG-110 graphite emissivity of 0.8 is conservative for air ingress simulations. Emissivity of 1.0 and 0.80 are suggested as the upper and lower bound respectively for sensitivity analysis.
- Correlations between normal spectral emissivity and temperature were developed for non-oxidized and oxidized IG-110 graphite, which are given as ($\lambda = 1.55\mu m$, 95% confidential interval):

$$\varepsilon_\lambda = (0.881 \pm 0.025) - (4.25 \pm 3.18) \times 10^{-5} T \quad (\text{non} - \text{oxidize})$$

$$\varepsilon_\lambda = (0.908 \pm 0.012) - (2.56 \pm 1.56) \times 10^{-5} T \qquad (\text{oxidized})$$

- According to the above correlations, at 1600˚C, which is the safety criteria of HTGR, the emissivity of non-oxidized and oxidized graphite are estimated to be 0.813 and 0.867 respectively.

RELAP5/SCDAP code modification: RELAP5/SCDAP code was improved for analysis of HTGR. A modification was made for the multiplication factor M(B) before code implementation. The improved RELAP5/SCDAP code was validated against several steady state and transient graphite oxidation experiments. Comparison between the code's prediction and experimental data leads to the following conclusions:

- The trends of gas mole fractions can be predicted by RELAP5/SCDAP. Specifically, the code predicts that as temperature increases, the graphite oxidation is in favor of generation CO, but as temperature further increases, the CO mole fraction starts to decreases due to CO combustion.
- The graphite oxidation model tends to over predict the reaction rate, which implies that it would give conservative prediction for an event of air ingress. Regime III is the high temperature regime where oxidation proceeds most rapidly and therefore it is of great importance in air ingress analysis. The deviation of simulation results from the experiment in regime III was within 20%, which is acceptable for a system analysis code like RELAP5/SCDAP.
- Compared to the original M(B) proposed by Kim, the graphite oxidation simulation preformed with the modified M(B) agreed better with graphite weight loss data. In other words, the accuracy of the graphite oxidation model in low temperature regime was improved.