Experimental determination of the emissivity of nuclear graphite at high temperature conditions

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Abstract
In a High-Temperature Gas-cooled Reactor (HTGR), radiation is the dominant form of heat transfer due to the high temperature environment. Therefore, the emissivity of the core materials (mainly nuclear grade graphite) is important for reactor safety assessment. In this paper, the emissivity of nuclear grade graphite IG-110 was measured in the temperature range from 500 °C to 1000 °C by using an infrared thermometer. Besides, the impact of the graphite oxidation, which may take place in a postulated air ingress accident, was also evaluated. As a result, it was found that the emissivity of IG-110 grade graphite decreases slightly as the temperature increase. Moreover, a relatively high emissivity was detected in the pre-oxidized specimen. Based on the measurement data, two experimental correlations were suggested for the engineering applications. It could also be concluded that the commonly used value of the graphite emissivity (0.8), is conservative for engineering judgment.

Keywords : Emissivity measurement, Nuclear graphite, Oxidation, Infrared thermometer, High-Temperature Gas-cooled Reactor

1. Introduction

High Temperature Gas-cooled Reactor (HTGR) technology has attracted increasing interest in many countries around the world in recent years because of its unique inherent and passive safety features. In most of HTGR, graphite is widely used because of its ideal material properties such as chemical inertness, high thermal conductivity, good corrosion resistance, and solid mechanical performance at elevated temperature. However, in a postulated air ingress accident, the graphite could chemically react with the invaded oxidant, which may lead to the degradation of its mechanical and thermal properties. Based on the previous researches, the kinetics of graphite oxidation could be derived as a function of the local temperature (Blyholder and Eyring, 1957; Cristian et al., 2008; Fuller and Okoh, 1997; Luo et al., 2004). Therefore, it is crucial that the heat balance of the system be studied for HTGR’s safety assessment.

In HTGR, radiation is of great importance to predict the heat balance since it is the dominant form of heat transfer due to the high temperature environment. To estimate the radiation heat transfer, the surface emissivity of the major core components (nuclear graphite) is required. In the reactor design, a value of 0.8 is commonly used as the graphite emissivity while predicting the radiation heat transfer between the fuel rods and the wall of the coolant channel (Fenech, 2013). However, as a dynamic property, emissivity varies with temperature and surface conditions. To this end, the objective of this study is to obtain the correlation between nuclear graphite’s emissivity and temperature with different oxidation degree.

Several studies have been performed on measuring the emissivity of various commercial graphite products at elevated temperatures since the 1950s (Grenis et al., 1962; Thorn and Simpson, 1953). Generally, the graphite emissivity was reported in the range from 0.95 to 0.70 (Pieson, 2012), and tends to decrease as the temperature increase (Null and Lozier, 1958). Specifically, Seo (2009a; 2009b; 2011) measured the emissivity of several types of nuclear graphite in the temperature range from 100 °C to 500°C and indicating a positive relationship between graphite emissivity and oxidation degree.
Nowadays, the method of the emissivity measurement could be classified into two types: the calorimetric method and the radiometric method. The calorimetric method measures the emissivity by evaluating radiation heat transfer of the system (Greene et al., 2000), while the radiometric method measures the emissivity of a specimen directly by using the specialized instruments, such as pyrometer (Wan et al., 1994), spectroscope (Krishna et al., 1999), radiometer (Iuchi et al., 2003) or Fourier Transform Infrared spectrometer (Zhang et al., 2003). In this study, a direct radiometric method is employed by using a single band infrared thermometer. The test specimen is made of IG-110 grade graphite, which is applied in both HTTR and HTR-10 (Only two alive HTGR reactors nowadays).

2. Experimental setup
2.1. Experiment apparatus

As shown in Fig. 1, the experimental apparatus consists of an infrared thermometer, a stainless steel chamber, a power supply system, a data acquisition system, a test specimen, and a K-type thermocouple. The internal surface of the chamber was coated with high-emissivity paint ($\varepsilon$: 0.94). An observation window, which was made of TEMPAX glass, was installed for temperature measurement.

![Fig. 1 Sketch of the experimental apparatus](image)

As shown in Fig. 2, the test specimen was mounted between two electrodes and was electrically heated by the power supply system. The temperature of the specimen was measured directly by a K-type thermocouple (accuracy +/- 5.5°C or +/- 1.0%), which was installed in a hole drilled at the center of the rear side. An infrared thermometer (IR-CZQH7T) is installed outside 40cm of the chamber to measure the temperature of the measuring point (same point as the thermocouple’s) through the observation window. IR-CZQH7T is capable in the temperature range from 500°C to 3500°C with an accuracy of +/- 5.0°C or +/- 0.5%.

As depicted in Fig. 3, the test specimen was machined into a rectangular bar of which the length, width, and height were 10mm, 10mm, and 30mm, respectively. An alignment mark was imprinted on the front surface to help the thermometer align the measuring point.

2.2 Experimental procedure

The experiment is divided into the pre-test and the main-test. The pre-test was performed firstly to confirm that the reliable readings could be obtained. The effect of the ambient conditions (e.g. the background radiation of the chamber, the presence of the observation window, or the presence of certain components in the atmosphere) would be evaluated and corrected in this stage.
The protocol of the pre-test can be summarized as follows:

1) The test specimen is coated with a matte black paint (Rust-Oleum MH21005) of which the emissivity equals to 0.94;
2) IR-CZQH7T is preheated for half an hour;
3) Argon gas is injected into the sealed chamber at a flow rate of 10.0 SLPM;
4) Power supply system is turned on once the oxygen concentration inside the chamber drops below 0.5%;
5) Once the K-type thermocouple’s reading reaches the target temperature point, quickly open the observation window, record the reading of infrared thermometer, then closed the window;
6) The temperature of the specimen is controlled back to 400°C;
7) The test specimen is heated again to a higher target temperature;
8) Steps 5 - 7 are repeated until the last target temperature is reached;
9) The temperature of the specimen is gradually decreased to 300°C, then the power was turned off.
10) The argon injection is turned off once the specimen’s temperature drops to 150°C.
11) Remove the chamber and repeat the steps 2 - 10.
12) Summarize the measurement results, and calculate the spectral emissivity based on the definition of spectral
radiance (Plank, 1914) with the following equation:

\[ E = \frac{e^{\frac{hc}{k_BT_k}} - 1}{e^{\frac{hc}{k_BT_{ir}}} - 1} \]  

(1)

where \( h \) is the Plank constant, \( c \) is the speed of light in vacuum, \( k_B \) is the Boltzmann constant, \( \lambda \) is the wavelength of the infrared thermometer, \( T_{ir} \) is the reading of the Infrared thermometer (representing the radiance of a black body) and \( T_k \) is the reading of the K-type thermocouple.

The experimental procedure of the main test is similar to the pretest (steps 2–10). The main experimental parameters of both pre-test and main-test are summarized in Table 1.

Table 1 Main experimental parameters of the pretest and main test

| Specimen                  | Pretest          | Main Test                  |
|---------------------------|------------------|----------------------------|
| Black paint coated        | As-received      | Pre-oxidized (Burn-off: 2.8%) |
| Temperature               | 600 °C–900 °C   | 500 °C–1000 °C             |

3. Results and Discussion

3.1 Effect of ambient conditions

The transmittance of a 5 mm TEMPAX glass (material of the observation window) is about 90.0% at a wavelength of 1.55μm (glass dictionary, 2017). In the pretest, the emissivity of a black paint coated specimen was measured with and without the observation window, respectively. As shown in Fig. 4, a transmittance of 89.3% is calculated by dividing the emissivity measured with the observation window by the one without. This value is pretty close to the one reported in the reference (glass dictionary, 2017).

Moreover, the emissivity measured in the test (without window) matched well with the value provided by the manufacturer of the black paint (0.93–0.94), which suggests that the environmental factors (e.g. the presence of air and argon on the transmission path, the radiation reflected from the internal surfaces of the chamber, etc.) are negligible in this experiment.

3.2 Measurement of IG-110 emissivity

As shown in Fig. 5, a total number of 53 measurements were obtained with the as-received graphite specimen, of which the maximum, minimum, and average values were 0.88, 0.81, and 0.85, respectively. It could be observed that the emissivity decreased slightly as the temperature increased. An experimental correlation was derived based on the experimental data, which is expressed as follows:

\[ \epsilon_{\lambda} = 0.881 - 4.25 \times 10^{-5}T \]  

(2)

where \( \epsilon \) and \( T (\circ C) \) are the emissivity and the temperature, respectively. The maximum deviation between the correlation prediction and the experimental data is 4.02%.

The emissivity of the pre-oxidized graphite specimen was measured by a similar procedure. As shown in Fig. 6, a total number of 54 measurements were obtained of which the maximum, minimum, and average values are 0.91, 0.87 and 0.89, respectively. Similar to the measurement from the previous tests, it was found that the emissivity decreased slightly as the temperature increase. The linearly-fitted correlation was as follows:

\[ \epsilon_{\lambda} = 0.908 - 2.56 \times 10^{-5}T \]  

(3)
It could be found that the emissivity increased approx. 4.8% after being oxidized, which is probably resulted by the enlarged emission area on the porous surface. In other words, although the graphite oxidation during the accident may lead to the degradation of the mechanical and thermal properties, it is favored for reactor cool-down because of the enhancement of the residual heat removal by radiation. Besides, it could be concluded that the value that the commonly used in the core design (ε: 0.8) is conservative for the safety analysis of HTGR. However, utilization of the empirical correlations derived in this study is recommended for a better estimation.
Fig. 6 Emissivity measured for pre-oxidized IG-110 graphite

4. Conclusions

A direct radiometric method is employed to measure the thermal emissivity of IG-110 grade graphite in the temperature range from 500 °C to 1000 °C. Based on the discussion on the results, the following conclusions could be drawn:

1) Based on the measurement data, the emissivity of IG-110 graphite could be derived as a function of temperature, which is given as:
   
   \[ \varepsilon_A = 0.881 - 4.25 \times 10^{-5} T \]

   Pre-oxidized graphite (2.8% burn-off):
   \[ \varepsilon_A = 0.908 - 2.56 \times 10^{-5} T \]

2) The oxidized specimen owns a higher emissivity because of the porous surface that generated during the graphite oxidation. In other words, although the graphite oxidation during the accident may lead to the degradation of the mechanical and thermal properties, it is favored for reactor cool-down because of the enhancement of the residual heat removal by radiation.

3) The emissivity value that commonly used is proved conservative according to the experiment results. However, utilization of the empirical correlations derived in this study is recommended for a better estimation.

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