Effects of Surface Roughness on the Emissivity due to Oxidation of Nuclear Graphite

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Abstract

The emissivity of the nuclear graphite was measured in comparison with the oxidation degrees. IG-11 nuclear graphite has been used as samples. Oxidation reaction of the graphite was carried out in air (5 L/min) using tube furnace (600℃; chemical reaction regime). Oxidized (6.15%, 10.05% weight loss) and non-oxidized samples have been chosen for the emissivity measurement. There were five different temperatures of 100℃, 200℃, 300℃, 400℃ and 500℃ for measuring emissivity. The emissivity of non-oxidized graphite increased when the temperature of measuring emissivity was above 200℃. The emissivity of oxidized graphite was decreased by the increase of temperature. The overall emissivity of oxidized sample of IG-11 is much higher than the other samples (0% weight loss). Differences in the emissivity behaviors of two samples were discussed in the scope of the differences in the surface area and surface roughness.

Keywords: nuclear graphite, emissivity, oxidation reaction.

1. Introduction

Graphite has good thermal and electric conductivity, and chemically very stable therefore it has been using for a variety of industrial matters. Especially Graphite for the nuclear reactor is used to the moderator, reflector and supporter in which fuel rod inside of nuclear reactor. Graphite has very strong endurance form neutron in comparison with other metals or ceramics therefore it has chosen for almost all nuclear reactor. When Graphite is exposed to neutron for a moment, it occurs to shrink and as long as it is exposed it starts to expand. Shrinkage of graphite in the nuclear reactor controls the stress from the contraction for the design of nuclear reactor but it is unsuitable for the controlling the stress form expansion, which is fatal problem for that. Therefore, durability of graphite is defined as a period between from contraction to expansion measured with neutron.

The graphite used in nuclear reactor has comprehension of endurance for neutron but because of the defect in the minute tissue and impurities, it reacts fatally so durability of graphite is reduced. The other fact that makes the durability of graphite is oxidation in which reacting with oxygen to CO_2. Graphite is basically occurring oxidation over 500°C in the air. VHTR is operated over 900°C which is necessary that the streaming gases in heat exchanger are used an inert gas such as Ar, He, for the prevention of graphite oxidation. However it is very important to take care of the specific characters of oxidation because of the reaction with an air-inflow or water as impurities in the inert gas. Therefore, for the nuclear reactor, it is highly recommended to use a high density and high purity of isotropic graphite. In this study, the emissivity of the nuclear graphite was measured in comparison with the oxidation degrees. IG-11 nuclear graphite has been used as samples and oxidized in air at 600°C. The surface roughness and crystallinity were measured to explain for the emissivity changes.

2 Experiments

2.1 Oxidation Process

The oxidation process for the IG-11 was carried out in air using tube furnace (600℃, chemical reaction regime). The flow rate of air was 5 ℓ/min and a round shaped quart used as a boat. Even though determined oxidation degrees were 5%, 10%, the results were 6.15% and 10.50% in the experiment.

2.2 Emissivity

Emissivity as a function of oxidation degree was measured through IR apparatus. The thermal heat form a black body and a sample are measured by the detector in order to measure the sample's emissivity standardized by a black body's emissivity as 1. The measuring conditions were 22℃ normal temperature and 35% humidity. There were 5 different measuring variables, 100℃, 200℃, 300℃, 400℃, and 500℃ for the sample of 0%, 6.155% and 10.50% burn-offed.

2.3 Raman Spectroscopy

Oxidized and original samples were analyzed and compared to Id/Ig using Raman spectroscopy (LabaRam HR, Jobin-Yvon, France). LabaRam HR system has 800m of focal distance which effect to the high limit of resolution and efficient accumulation of scattering light. It also has advantage in extremely small field such as micrometer measured by confocal system.

2.3 Surface Morphology

To observe the morphology changes between before and after oxidation of samples, scanning electron microscope (SEM) were used. For the SEM observation, samples were 60° tilted to get higher brightness. Surface morphologies were observed without polishing.

2.4 Surface Roughness

Surface roughness of samples were measured by α -step apparatus (Dektak 6M). The maximum measuring depth from the sample surface of the apparatus was 250 ㎛.

3. Results and Discussions

3.1 Emissivity of Nuclear Graphite

Table1 and Figure1 show emissivity results. The emission power increases with high oxidation degree and high temperature measured. However, Emissivity shows different, first of all, 0%wt. loss sample is decreasing its emissivity between 100℃ to 200℃ and increasing from 200℃. It totally has lower emissivity than samples are oxidized. The 6.15%wt. loss sample and 10.50%wt. loss sample are decreasing its emissivity during the measuring temperature is going up. It has higher emissivity than non-oxidized sample. Figure 1 refers to variation of emissivity by variation of high oxidation rate and a function of measuring temperature. High oxidation degree expects high emissivity because of the surface roughed. The emissivity is decreasing when the temperature is increasing.

Table 1. Result of the emissivity measurement.

Samples	Temperature (℃)	Emission Power (W/㎡)	Emissivity (3~20 ㎛)
IG-11 0% wt. loss	100	6.130x102	0.712
	200	15.192x102	0.599
	300	33.906x102	0.611
	400	64.808x102	0.626
	500	120.296x102	0.650
IG-11 6.15% wt. loss	100	7.055x102	0.819
	200	19.264x102	0.760
	300	41.926x102	0.755
	400	77.233x102	0.746
	500	128.260x102	0.693
IG-11 10.05% wt. loss	100	7.220x102	0.839
	200	19.305x102	0.762
	300	42.171x102	0.760
	400	78.232x102	0.756
	500	137.829x102	0.729

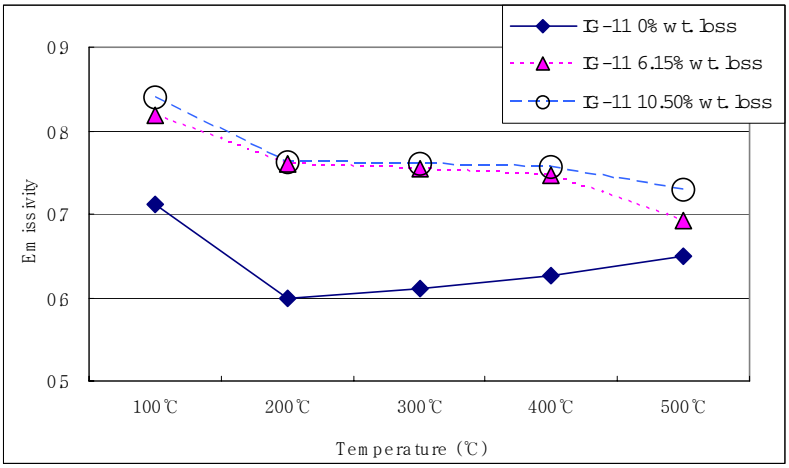

Figure 1. Emissivity changes as a function of measuring temperature.

3.2 Crystallinity

Table 2 is a result of Raman analysis. *Id/Ig* result of oxidized sample is measured smaller than 0% wt. loss sample and average *Id/Ig* result for 6.15% sample and 10.50% sample are almost same. It says when sample is oxidizing, the amorphous substance is burn-offed first.

Table 2. Summary of Raman analysis

Sample	#1 (*Id/Ig*)	#2 (*Id/Ig*)	#3 (*Id/Ig*)	#4 (*Id/Ig*)	#5 (*Id/Ig*)	Average (*Id/Ig*)
0%	0.37	0.21	0.12	0.24	0.14	0.22
6.15%	0.09	0.22	0.25	0.16	0.09	0.16
10.50%	0.23	0.12	0.06	0.13	0.19	0.15

3.3 Surface Morphology Changes

According to figure 2, surface of original sample (0%) keeps much smooth surface than oxidized samples (6.15% and 10.50%). The smooth surface of irregularity is increasing when the oxidation is progressing. It means that the pore is not only oxidized fast but also disordered phase oxidation, which is organized nearby cokes, is fast too. Therefore, it is assumed that the large pore which easy to observe is not only well developed, but also the small pore which can't observe is well developed as well. Figure 3 is the enlarged photo of surface structure which certainly observe.

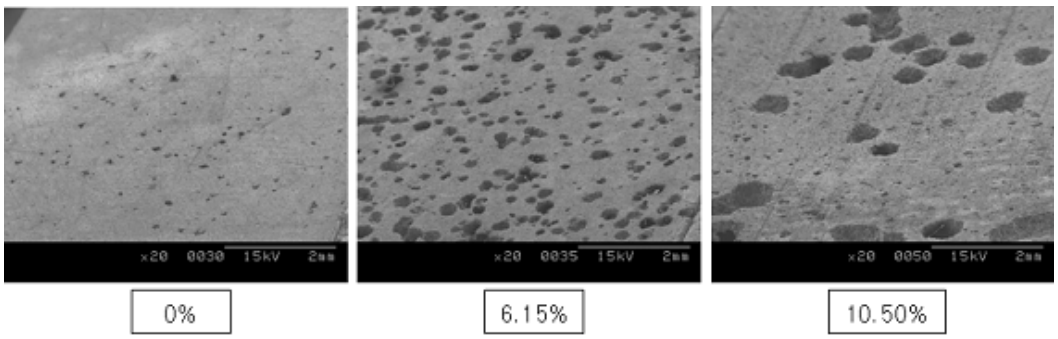

Figure 2. Surface morphologies of IG-11 (60° tilted, ×50).

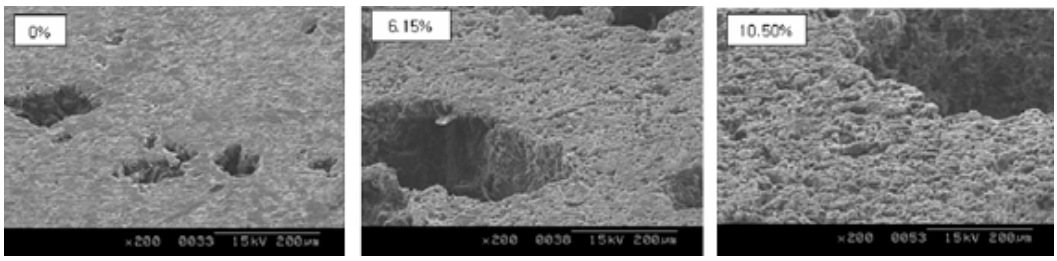

Figure 3. Surface morphologies of IG-11 (60° tilted, ×200).

3.4 Surface Roughness

Figure 4 shows surface roughness profiles of IG-11s. The surface roughness increases with oxidation degrees. The 6.15% wt. loss sample has large pore in the range of 100 to 2500 ㎛ depth and around 500 ㎛ width. The 6.15% wt. loss sample has huge pore over the detecting range over 2500 ㎛ depth and around 1000 ㎛ width as shown in SEM results. The average surface roughness (Ra) of relatively smooth surface, except large pore area, is listed in Table 3. It should be calculated quantitatively between the surface roughness and the emissivity.

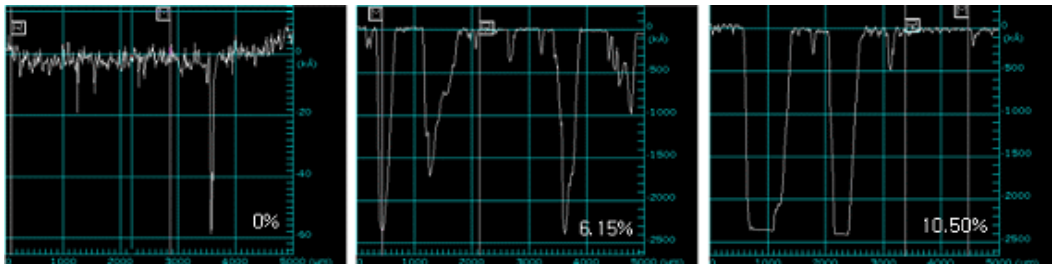

Figure 4. Surface roughness profiles of IG-11s.

Table 3. Summary of Raman analysis

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10.50%	0.23	0.12	0.06	0.13	0.19	0.15

4. Conclusions

The emissivity as a function of oxidation degree for the IG-11 and the surface roughness and crystallinity are estimated to serve explanation of the emissivity changes. The emissivity is increasing after oxidation over 6.15% wt. loss. The 6.15% wt. loss and 10.05% wt. loss show little difference for the emissivity. The surface roughness is increasing after oxidation and the pore size is increasing as well. The crystallinity shows similar inclination as the emissivity, there is little difference of crystallinity in the sample more than 6.15%wt. loss.

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