# Oxidation and Temperature Effects on the Fracture Toughness of Nuclear Graphite at VHTR.

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### 1. Introduction

Understanding the effects of oxidation and temperature of nuclear graphite are important for the design and safety analysis of the very high temperature gas-cooled reactor (VHTR), because the graphite experiences structural degradation via oxidation during normal reaction operation or accident. At present, the most critical event expected in this type of reactor is an air-ingress accident, which is caused by a pipe break. When air is ingressed into the reactor due to an accident, the graphite materials in the moderator and reflector suffer from a chemical reaction with oxygen [1]. Until recently, several researchers have investigated the effect of oxidation on mechanical properties of nuclear graphite [2-3]. For example, a 50% tensile strength reduction by about 10% weight loss was reported. In the present study, we tested and estimated the fracture toughness of nuclear graphite in various temperature  $(25^{\circ}C \sim 700^{\circ}C)$  after uniform oxidation by 0 to 15%.

#### 2. Experimental

Four different nuclear graphite were investigated; IG-110, IG-430 isostatically molded and purified by Toyo Tanso Co, Ltd and NBG-18, NBG-17 vibrationally molded purified by SGL Carbon Co, Ltd. Physical properties of graphite are summarized in Table I. Single edge notched beam (SENB) specimens were machined from graphite blocks. The fracture toughness values were measured by 3-point bending test at the room temperature, 600°C, and 700°C in flowing nitrogen environment (10 L/min) for the pre-oxidized specimens. The dimension of the SENB specimen was 100 mm in length (L), 80 mm in the distance between support spans (S), 4 mm in initial crack length (a<sub>0</sub>), 10 mm in width (W).

Table I: Typical physical properties of nuclear graphite (from manufacturer's data)

Manufactur er	Toyo Tanso		SGL	
Grade	IG-110	IG-430	NBG-17	NBG-18
Coke	Petroleum	Pitch	Pitch	Pitch
Fabrication	Isomolded	Isomolded	Vibro- molded	Vibro- molded
d <sub>avg</sub> (μm)	20	10	80	160

#### 3. Results and discussion

#### 3.1 Fracture toughness at high temperature

Fig. 1 shows fracture toughness vs. temperature curves obtained by 3-point bending test. Fracture toughness of all graphite is increased by thermal expansion of grain at high temperature. It was reported that coefficient of thermal expansion (CTE) increases from  $2x10^{-6}$  to  $7-8x10^{-6}$  as the grain size decrease from 3000 to  $1\mu$ m due to the better contact between neighboring particles of the material [4]. And increase rate of fracture toughness on fine grain graphite is higher than coarse grain graphite at high temperature, because CTE of fine grain is larger than coarse grain. Comparing the same manufacturer, increase rate of IG-430 is higher than IG-110 and increase rate of NBG-17 is higher than NBG-18.



Fig. 1. Fracture toughness vs. temperature curves of nuclear graphite (IG-110, IG-430, NBG-18, and NBG-17)

#### 3.2 Oxidation effect on the fracture toughness

Generally, fracture toughness of graphite is exponentially decreased with porosity [5]. And the porosity of the graphite increases with oxidation. Fig. 2 shows fracture toughness vs. weight loss curves is obtained by 3-point bending test. Normalized fracture toughness of nuclear graphite showed exponentially decreasing trend with oxidation. As the graphite is oxidized, the weakening effects on fracture toughness are caused by the proximity of pore due to increase in pore size or creation of pore. Crack can jump from one pore to the next pore [6]. Comparing the same manufacturer, decrease rate of IG-110 is a little higher than IG-430. As the grain size of IG-110 has a little bigger than IG-430, primary crack of oxidized IG-110 can propagate easily than IG-430. However, as the difference of grain size both materials is not big, normalized fracture toughness of IG grade is similar. In case of NBG grade, test result significantly shows difference of fracture toughness at same condition. Decrease rate of NBG-18 is higher than NBG-17. The difference of grain size affects the fracture toughness of oxidized graphite. Decrease rate in the fracture toughness of the oxidized graphite is faster in NBG-18 than in the other graphite.



Fig. 2. Effect of weight loss on fracture toughness (IG-110, IG-430, NBG-18, and NBG-17)

## 3.3 High temperature effect on fracture toughness of oxidized graphite

Fracture toughness of nuclear graphite is increased at high temperature and decreased with weight loss. In this section, we considered the effects of oxidation and temperature together. Fig. 3 shows fracture toughness vs. weight loss curves obtained by 3-point bending tests. In the initial weight loss period, the fracture toughness of all graphite is higher at high temperature (600°C, 700°C) than at the room temperature. Since the fracture toughness is increased by thermal expansion of the grain at high temperature, increasing effect of fracture toughness shows up at a little oxidation level. Increasing effect is called "High temperature effect". As high temperature effect is higher than oxidation effect at low oxidation region, fracture toughness of nuclear graphite is dominated by this effect. However, the high temperature effect with the weight loss is decreased by accommodation of thermal expansion of pores. In addition, the compressive stress in the binder phase is released by accommodation of the thermal expansion.

Over 10% oxidation, the fracture toughness of nuclear graphite at high temperature is almost same value at room temperature. As the high temperature effect is almost disappeared at high oxidation region, fracture toughness of nuclear graphite is controlled by oxidation effect. From our test results, although graphite is oxidized about 2%, the fracture toughness is maintained by high temperature effect.



Fig. 3. Effect of high temperature effect on fracture toughness (IG-110, IG-430, NBG-18, and NBG-17)

#### 4. Conclusions

Temperature and oxidation effects on the fracture toughness of nuclear graphite were investigated. Grain size of the coke particle shall affect the fracture toughness of the nuclear graphite. Comparing the same manufacturer, the difference in grain size affects the increase rate of fracture toughness with temperature. Increase rate of fracture IG-430 is higher than IG-110, and NBG-17 is higher than NBG-18 at 600°C.

Fracture toughness of nuclear graphite showed exponentially decreasing trend with oxidation. As the proximity of pore makes primary crack to jump one pore to next pore as oxidation progresses, normalized fracture toughness is exponentially decreased by weakening effect. Comparing the same manufacturer, decrease rate of IG-110 is a little higher than IG-430, and decrease rate of NBG-18 is higher than NBG-17. High temperature effect is higher than oxidation effect at low oxidation region. However, this effect is disappeared by accommodation of thermal expansion at high oxidation region.

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