Transactions of the Korean Nuclear Society Spring Meeting
Chuncheon, Korea, May 25-26 2006

Effects of specimen size, ingredient filler and pores on the specific electrical resistance (SER) of nuclear graphite

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1. Introduction

It is well known that the specific electrical resistivity (SER) of materials, including the nuclear graphite, changes with the raw materials and manufacturing process. In the standard nuclear graphite specification (draft) [1], the SER is used for the estimation of the graphitization temperature or for the prediction of the degree of graphitization. All of these applications of SER are based on the strong correlation between the SER and the microstructure of the materials, i.e., for example, degree of perfection of crystal lattice, impurities, grain size and grain boundaries, or pore structures.

For its high applicability, however, the current Standard Test Method for Electrical Resistivity of Manufactured Carbon and Graphite Articles at Room temperature, ASTM C 611-98, lacks information on the optimum size of specimens by specifying only the ratio of specimen length to maximum cross-sectional dimension (width or diameter) as 6:1 [2].

In this work, the effects of specimen size on the SER were investigated for a range of specimen cross-section and the results were compared with respect to the microstructure (crystallite size, pore) of the candidate graphites in view of graphite selection.

2. Experimental

Specimens with various cross-sections, i.e., (1×1) to (10 ×10) mm^2, were prepared from the isotropic nuclear graphites IG-110, IG-430 (Toyo Tanso, Japan) and NBG-18, NBG-25 (SGL, Germany). The minimal ratio of the specimen length to width was 7:1. After machining, all the specimens were cleaned in acetone filled ultrasonic bath. To remove all the possible humidity from the specimens, all specimens were annealed at 110 ^{0}C after drying 2 hours in air.

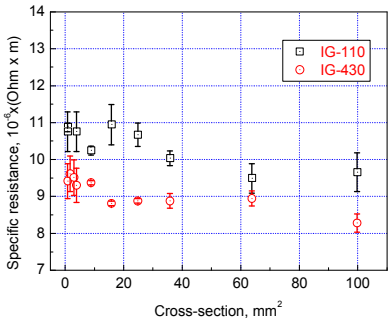
The method of a DC drop potential measurement was used for SER measurements. Nanovolmeter 2182A (KEITHLEY) was used for drop potential measurements.

For open porosity measurement, the helium gas pycnometer (model AccuPyc 1330, Micromeritics) was used. For the crystallite size, L_a,, determination, Raman intensities were measured. The L_a was determined from the ratio of the intensity of D and G peaks (I_D/I_G) [3], i.e., $\frac{I_D}{I_G} = \frac{A}{L_a}$, where, A = 44 (Å).....(1)

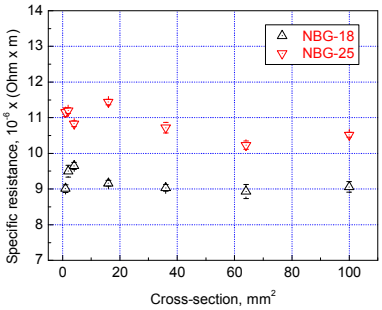
3. Results and discussion

3.1. Effect of specimen size and Coke

Figure 1 shows the specimen size effects, i.e. cross-section of the specimen, on the SER. If the large scattered SER data on the small cross-section area (below about 20 ~ 30 mm^2) are excluded, all the SER data show a relatively constant value up to 100 mm^2. Thus, with the increase in the specimen size, both the IG-110 and NBG-25 of petroleum coke show a SER of 9.5 ~ 10.5 x 10^{-6} (Ohm.m), and the IG-430 and NBG-18 of pitch coke show a SER of 8.3 ~ 9.0 (Ohm.m).



(a)



(b)

Figure 1. Changes of SER with the specimen size (cross section) and graphite grades.

One of the reason for the lower SER of pitch coke graphite may be attributed to its higher density (i.e., lower porosity in the Table 2) than the petroleum coke graphite.

3.2. Scattering of carriers on crystallite boundaries

When the sizes of crystallites are of a few hundred angstrom, it is known that the crystallites boundary scattering play a noticeable role at low temperature [4,

5]. In view of this observation, in Table 1, the smaller crystallite sizes of the pitch coke graphites (IG-430 and NBG-18) should show a lager SER than the petro coke of a larger coke size. However, as seen in the Figure 1, the results of SER contradict to this prediction. Reasons for this observation are not clear at the moment, however, that further investigation is required to explain the current SER-La correlation considering the Raman spectroscopy testing temperature and the prediction of La by equation (1).

Table 1. The intensity of ratio (ID/IG) and crystallite sizes of graphite.

Graphite	I_D/I_G	$L_{a,}$ Å
IG-110	0.12	367
IG-430	0.16	275
NBG-18	0.23	191
NBG-25	0.15	293

3.3. Effect of porosity on SER.

To investigate the porosity effects on the SER, total density, picnometric density d_P, total porosity, and closed porosity were determined. An apparent total porosity ε of graphite can be determined from the apparent density d by

$$\varepsilon = \frac{(2.25 - d)}{2.25} \quad \text{.....(2)}$$

The value 2.25 g/cm^3 is the density for a single crystal graphite. The open porosity ε_0 was determined from the picnometric density d_P. The results of porosity measure- ments and calculations are summarized in Table 2.

Starting from the experimental data the closed porosity ε_C was calculated as a difference between the total and the open porosity. It is evident that the total porosity depends also from the density of filler and binder.

Table 2 shows that the total porosity of the pitch coke graphite, i.e., IG-430 and NBG-18, is smaller than the petroleum coke graphite. This observation predicts well the results of SER determination. Thus, it is predicted that the SER of the pitch cokes will be smaller than the petroleum coke graphite, i.e., IG-110 and NBG-25 as we observed in the Fig. 1, possibly due to the higher density of pitch coke graphite (i.e., lower porosity density of pitch coke). From Figure 1 and Table 2, the strong dependency of SER on the porosity, rather than the dependency on the crystal size, is confirmed.

Further, this observation may be confirmed in Figure 1 where, the IG-430 and NBG-18 of pitch coke graphite show an early stabilization of SER by the relatively smaller pore density.

Table 2. Determination of the porosity by an experiment or calculation.

	IG-110	IG-430	NBG-18	NBG-25
d, g/cm^3	1.77	1.82	1.853	1.82
d_P, g/cm^3	2.0541	2.1023	2.0149	2.0751
ε, %	21.33	19.11	17.644	19.11
ε_0, %	13.83	13.43	8.035	12.29
ε_C, %	7.5-8.71	5.68-6.56	9.6–10.45	6.82–7.77

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4. Conclusion

(1) Minimum cross-section of the specimens for the SER measurement of the isotropic nuclear graphite appeared as 40 mm^2.

(2) The pore density dependency of SER was confirmed. The higher the density, the lower the SER (Petroleum based graphite: 9.5 ~ 10.5 x 10^{-6} (Ohm.m), Pitch: 8.3 ~ 9.0 (Ohm.m)).

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