

## **Title: Highly porous expanded graphite: thermal shock vs. programmable heating**

**Manuscript ID: materials-1470412**

*Bannov et al.* describes a new method to produce highly porous expanded graphite from intercalated graphite. The article related a method known as programmable heating, at different temperatures ranged from 400 to 700 °C, that is compared with thermal shock method at 400 and 1000 °C.

The expanded graphite at 400-500 °C display the best characteristics in terms of bulk density, high surface area and oxidation resistance making these materials suitable for catalyst and electrochemical applications.

In my opinion this article could be of interest of readers of Materials, but the article is plenty of small mistakes which have to be corrected before I can recommend for its publication. In addition, some points have also to be clarified. My comments are listed below:

### **Abstract:**

#### **Page 1, line 21**

“678 and 593 m<sup>2</sup>/g”

Please check de values in accordance with the data of table 3 and correct.

### **Introduction:**

#### **Page 1, line 33.**

Could you please introduce a reference related to catalyst applications?

#### **Page 1, line 35**

Can your briefly explain the liquid-phase-expansion method?

#### **Page 1, line 42**

Can your briefly explain the features of EG obtained by FeCl<sub>3</sub> gas phase intercalation method?

#### **Page 2, lines 49-51**

“Goudarzi et al. [26] noted that the particle size of GICs and temperature of exfoliation significantly affected the pore size of expanded graphite. Textural characteristics increased when the temperature increased from 700 to 900 °C.”

The influence of particle size is missing in the text. Please include.

#### **Page 2, line 57**

“(graphite bisulfate)”. This information should be removed from the introduction section and added to the experimental section.

#### **Page 2, line 58**

You made a comparison between thermal shock and programmable heating, which should be indicated in the introduction.

## **2. Experimental**

### **Page 2, lines 71-83**

Please indicate the atmosphere conditions for programmable heating and thermal shock. I suppose is air atmosphere.

In addition, why you don't use the same temperatures for both programmable heating and thermal shock in order to make a reliable comparison? In fact, the best conditions for programmable heating are obtained at 500 °C, so it would be recommended to employ the same temperature for the thermal shock experimental conditions.

### **Page 3, line 86**

Figure 1 can be move to supplementary information. In addition, the scale bare should be included.

## **2.2. Characterization**

### **Page 3, lines 71-83**

If possible, please complete the information related to the equipment's, indicating the trade mark and the country of each one.

### **Page 3, lines 100-101**

Please include the reference of Equation 1 and indicate how  $L_c$  was calculated. In addition, indicate the units.

## **3. Results and discussion**

### **Page 4, lines 128-137**

In the case you used air atmosphere for the thermal treatments, both for programmable heating and thermal shock, please explain why you use Ar atmosphere instead of air for TG/DSC experiments?

In addition, if you observed the WL continues at temperatures higher than 700 °C, please indicate why you haven't carried out the experiments at higher temperatures? Please also notice you employed higher temperature for the thermal shock experiments.

Please add some reference that confirm the exfoliation of intercalated graphite occurs at 267 °C (**page 4, line 130**).

### **Page 4, line 120**

Why do you use argon to determine the temperature of exfoliation when you used air to perform it experimentally?

### **Page 4, line 121**

Why do you use a mixture of argon and oxygen to determine the stability of samples against oxidation?

### **Page 4, line 139**

Please include (c,d and e,f, respectively).

### **Page 4, line 139**

Please check that “TEG1-400” is correct and change if necessary.

**Page 4, line 139**

Please, check the value “O (11.06 at.%)” with the one that appears in Table 2 (page 5, 1.06) and correct the wrong one.

**Page 6, line 184 and page 7, line 186**

Bulk density is not included in Table 3, please correct.

**Page 7, lines 192-196**

“The highest yield was reached at 400 °C, whereas the bulk density was not relatively high, which is inappropriate for some applications (adsorption, catalysis) where this value must be lower. Therefore, in order to use EG in these fields, the optimal yield and bulk density will be at 500 °C.”

Please explain why the bulk density must be lower for adsorption and catalysis applications.

**Page 7, lines 197-207**

The adsorption-desorption N<sub>2</sub> isotherms and pore size distribution must be included and discussed.

In addition, the authors should explain, and include in the experimental section, the methods and approximations made for the calculation of total surface area, surface area of micro and mesopores, total pore volume and average pore diameter.

**Line 198:** Please check “678 m<sup>2</sup>/g” and correct.

**Lines 200-203:** “The samples obtained using programmable heating and thermal shock were predominantly mesoporous, with a fraction of micropores within 22-27%, and for TEG1-400, TEG1-500, TEG1-600, and TEG1-700 it was 26.8, 26.0, 25.6, and 21.8%, respectively. Thus, increasing temperature induces a decreasing fraction of mesopores.”

I am so sorry but I don't understand the sentence, please explain better. From table 3 it seems clear that as the programmed temperature increases both micro and mesopores decrease. In contrast, the opposite occurs for thermal shock treatment.

**Lines 206-207:** “The differences in porous structure of the samples can be seen in SEM images shown in Figures 3 and 5.”

Please explain the differences found better.

**Page 8, Line 226**

Reference [34] is missing, please include.

**Page 8, line 233**

Please indicate how do you calculate L<sub>c</sub>.

Please if possible use same units nm or Å for the figure 7 and along the text.

**Page 9, line 238**

Please check “678 m<sup>2</sup>/g” and correct.

**Page 10, lines 259-261**

“The difference of I(D)/I(G) ratio of samples TEG1-400 and TEG2-400 obtained using programmable heating and thermal shock is high enough (0.1 and 0.014, respectively) and it is in agreement with textural characteristics shown in Table 3.”

I am so sorry but I don't understand the relationship of I(D)/I(G) values with the textural characteristics. Could you explain better?

In addition, could you check the I(D)/I(G) ratio of TEG2-400 sample?

**Page 10, Lines 265-267**

Why don't appear the C=S bonds in C1s XPS spectra at least for intercalated graphite sample? Please explain.

**Page 11, Line 277**

The O1s XPS spectra are missing. Please include.

**Page 11, Line 303**

“TEG1-400–TEG1-500 samples are shown in Figure 11.”

Please check and correct the sentence if necessary.

**4. Conclusions**

**Page 13, line 354**

“678 and 593 m<sup>2</sup>/g”

Please check the values in accordance with the data of table 3 and correct.

**Additional points:**

“Arb. Un” should be changed by arb. unit in figure 6(a and b) and 9 (and b) and added in figure 10(a,b).