Graphite surface profile with different polishing treatment

B A Budiman¹,²,*, P Sambegoro¹,² and P N Halimah¹,²

¹Faculty of Mechanical and Aerospace Engineering, Institut Teknologi Bandung, Indonesia
²National Center for Sustainable Transportation Technology, Indonesia

*bentang@ftmd.itb.ac.id

Abstract. Graphite is a fascinating material to study in nanotechnology. Surface profile and roughness of the graphite are some of the most important parameters to determine many physical and mechanical behaviors including friction or bonding condition. To acquire certain surface profile, mechanical polishing treatment is one of the easiest ways that can be applied. In this work, graphite is polished by three different grades of sandpapers (P800, P400, and P220) and cellulose-based paper. The surface properties are observed by a laser microscope and various surface roughness parameters were quantified. The sequential polishing process employed in this work is able to modify the surface roughness and might improve the physical properties. This work sheds light on the importance of the mechanical surface treatment on graphite.

1. Introduction

Graphite is multilayers of graphene, i.e. a hexagonally arranged carbon atoms in a planar condensed ring system [1]. These geometrical properties make graphite become one of the mineral products that have versatile and unique characteristics. The development of manufactured graphite began when synthetic graphite can firstly be produced using electrical resistance furnace [2]. More recently, natural graphite plays an essential role, such as high-performance structures for the linings in steel manufacture, higher charge capacity in lithium-ion batteries compared to other elements, and fiber’s material in graphite-based composites [3]. Graphite is also used for medicine because it is compatible with the human body for a long period [4].

Most of the graphite’s applications need specific surface characteristics which must be controlled well. Typically, the surface characteristics can be represented by roughness, waviness, and configuration or profile parameters [5]. The evaluation of those parameters is very important for many fundamental problems such as contact between surfaces, which could affect the friction coefficient, the tightness of contact joints, or the heat and electric-current conduction [6]. Recently, major studies of the surface characteristics in graphite are related to a modification of mechanical properties of graphite-based composite materials and improvement of battery’s anodes’ transfer electron capability.

In graphite-based composites, the graphite surface roughness directly corresponds to mechanical locking of an interface between graphite-based fiber and the matrix. This locking significantly contributes to the composites’ performance as load-bearing structures [7]. To obtain effective mechanical locking, the surface of graphite-based fiber must be controlled carefully. Some modifications of the surface roughness might also be conducted to optimize the interface strength which directly improves the composite performance [8]. However, controlling the surface roughness might not
be an easy task since the relationship between surface treatments with roughness parameters in the graphite has not been intensively studied yet.

Another utilization of graphite is driven by the growth of lithium-ion batteries’ manufacture. Natural graphite has so many benefits to be used in lithium-ion batteries because of the high density of electrical storage and battery charge cycling characteristics compared to other materials. Yoshio et al. reported remarkable improvements in the electrochemical performance of natural graphite after the TVD-carbon coating treatments, such as higher rate capacity, higher coulombic efficiency, lower irreversible capacity, and lower production cost [9]. A recent study by Mundszerger et al., showed that rotational impact blending of natural graphite into spherical shapes could reduce the surface area of graphite flakes which then could increase the anode packing density [10]. Further improvement of anode technologies in reversible charge capacity and cycling performance is presented by Zhang et al. by coating processed natural graphite with silicon nanoparticles [11].

Although it seems those studies quite practical to analyze the influence of graphite surface in the anode, there are some challenges too. First, a good result would need quite many surface parameters. The surface characteristics do not only depend on its roughness, but also its profile. Therefore, an extra effort is required to obtain and process the data, as conducted by Petrik et al. [12]. They use two tools, i.e. Atomic Force Microscopy (ATM) and Spectroscopic Ellipsometry (SE), to analyze the surface roughness. This research reported that surfaces having identical roughness parameter such as arithmetic average height ($R_a$) could yield different results because of the different FFT spectra. The second challenge is, it includes complicated work in manufacturing certain materials to meet certain roughness parameters. Researchers investigated some methods to predict the surface roughness after machining, such as numerical simulation [13] and optimizing the cutting parameters [14]. The optimized machining operation is needed to control the quality of the finished surface because the maximum height roughness parameter ($R_p$) depends significantly on the rotational cutting speed and workpiece diameter.

In this work, precise measurement has been applied in a small surface area of 5 graphite specimens to determine the quantitative values of surface roughness parameters. Comparison of roughness factors by three different grades of sandpapers (P800, P400, and P220) and cellulose-based paper by laser microscope has also been made. Furthermore, the effects of those treatments on the graphite surface profile are also comprehensively discussed.

2. Method

2.1. Specimen preparation

Specimens used in this work is cylindrical graphite with a diameter of 0.9 mm. Those specimens are treated differently i.e. unpolished and polished by cellulose-based paper, P800, P400, and P220 sandpapers). During the process, the surface profile of the graphite specimen was observed in a microscope to assure all surface region has been polished. After the uniform profile obtained, the surface roughness of those specimens is ready to be measured by using the microscope.

2.2. Observation of graphite profile and roughness

To obtain an accurate observation, a laser microscope Keyence VK 9710 was used. The microscope can clearly capture the graphite surface profile. The height and depth of the surface profile can be obtained easily. The microscope can also measure several surface roughness parameters, such as $R_a$, root mean square roughness ($R_q$), $R_p$, and a maximum depth of valley ($R_v$). Furthermore, the Amplitude Density Function (ADF) and Bearing Area Curve (BAC) curves can be analyzed from the microscope observation. In reference [6], those parameters are described in detail.

$R_a$ is the most commonly used surface parameters. $R_a$ measures the average absolute value of the surface irregularities, which consists of peaks and valleys. $R_q$ uses statistical methods to measure the standard deviation of the surface roughness’ distribution. $R_p$ is the value of maximum height between
all peaks above the mean line in the entire surface profile. The last parameter i.e. $R_v$ is the value of maximum depth between all valleys below the mean line in the entire surface profile. ADF is the histogram representation of profile heights’ distribution. ADF can be obtained by plotting the amount of identical profile height values on the x-axis and the value of profile height on the y-axis. One of ADF advantages is it can distinguish two profiles which have the same $R_a$ or $R_q$ value but has different surface shapes by observing the skewness parameter. The skewness parameter can be obtained by plotting the median line in the ADF curve. There are three skewness parameters, those are positive skewness, zero skewness, and negative skewness. Zero skewness means symmetrical height distribution in which a surface has peaks as many as valleys. Profiles with more points below the median line have positive skewness. Meanwhile, profiles with more points above the median line have negative skewness. The illustration of ADF curve and skewness parameter’s comparison between two different profiles having the same $R_a$ value is shown in Figure 1a. BAC is the representation of a solid profile percentage at a certain height. BAC can be obtained by plotting the measure of bearing line length on the x-axis and the value of profile heights on the y-axis. BAC can be used for the analysis of the load carrying surfaces because it represents the effective contact area of the surface. The illustration of BAC curve is shown in Figure 1b.

**Figure 1.** (a) The ADF and skewness parameter of two different surface profiles. Although both profiles have the same $R_a$ value, those have different surface shapes. (b) Profile B has a smoother surface. Thus, it has less effective contact area.

### 3. Results and discussion

Figure 2 shows surface profiles of cylindrical graphites with different polishing treatments. It can be seen that unpolished graphite originally has profile of continuous peak along with longitudinal direction. After polished by cellulose-based paper, the surface becomes smoother with the continuous peak profile is eliminated. Polishing by sandpapers can create scattered profiles with different surface roughness depending on the sandpaper types. Rougher surface profiles keep appearing as the sandpapers get rougher. It is also observed that polishing the graphite with P220 sandpaper create more nonuniform roughness than P400 and P800 sandpapers. These typical surface roughness and profiles determine the performance of the graphite in many engineering applications. Thus, it has to be quantified by using the surface parameters.

**Figure 2.** Five graphite specimens: (a) before surface treatments/unpolished specimen, and after surface treatments by: (b) cellulose-based paper, (c) P800, (d) P400, and (e) P220 sandpapers.
Table 1 lists the value of surface roughness parameters i.e. $R_s$, $R_q$, $R_p$, and $R_v$ from five graphite specimens. After observing the value of all parameters for each specimen, it is noticeable that graphite specimen after cellulose-based paper treatment has the lowest value of surface roughness parameters amongst all. Polishing the graphite with P800, P400, and P220 sandpapers create rougher surface than the original one. These values are predicted by considering visual observation as shown in Figure 2. However, all of these parameters still do not indicate surface profile difference among specimens.

**Table 1. Surface roughness parameters data of five graphite specimens.**

| Specimen no. | Surface treatment | $R_s$ | $R_q$ | $R_p$ | $R_v$ |
|--------------|-------------------|-------|-------|-------|-------|
| 1            | Unpolished        | 4.32  | 5.42  | 13.03 | 14.32 |
| 2            | Cellulose-based paper | 2.55  | 3.21  | 9.35  | 10.34 |
| 3            | P800              | 5.92  | 7.95  | 32.29 | 24.71 |
| 4            | P400              | 7.99  | 10.63 | 37.65 | 32.71 |
| 5            | P220              | 12.43 | 16.27 | 40.49 | 52.3  |

To analyze the difference of surface profile among specimens further, ADF curve is also investigated (see Figure 3a). It can be seen that the five specimens have different skewness which depends on the surface treatment. Without polishing treatment, the graphite has zero skewness which means there is a fairly balanced amount between the peaks and valleys. It can also be seen that there is an increase in the number of highest peaks and low valleys, indicating unpolished surface profiles have many high peaks and deep valleys. This analysis fits with visual observation shown in Figure 2a in which the unpolished graphite surface forms continuous peaks and valleys rather than a scattered profile. Similar ADF curve also appears for graphite surface polished by P220 sandpaper. The nonuniform roughness shown in Figure 2e can be captured by ADF curve with an irregular number of peaks and valleys. Thus, ADF curve can indicate whether the surface has certain profile or not.

The change in the ADF curve from unpolished to polished surface by cellulose-based paper illustrates the finer surface. After polished by the paper, it can be seen that height distribution becomes small indicating the loss of a high number of peaks and deep valleys. The ADF curve is also well distributed with mean value almost coincides with median value (zero skewness). In this case, the continuous peaks/valleys profile have been removed as observed in Figure 2b. Polishing with P800 and P400 also shows the same trend in which the height number is well distributed. The skewness of these ADF curve slightly becomes positive. The height variation in the graphite surface is wider because the emery of P800 and P400 sandpapers have larger particles than cellulose-based paper. Even though they can reduce the number of peaks and valleys, new peaks and valleys with more depth appears in small amounts. In addition, the result of a much rougher surface profile is obtained after graphite specimen is polished by P220 sandpaper. Negative skewness is also obtained for this case.

In designing surface that can bring good mechanical locking at graphite-based composite, zero skewness is required because the matrix can penetrate to the valley at the graphite surface properly. Negative skewness of the graphite surface potentially causes air bubble trapped in the valley which can initiate a crack. Thus, the interface strength due to mechanical locking might be low. In contrast, positive skewness can create ineffective mechanical locking since the small region of graphite penetrates to matrix does not contribute to the locking. This can ease the crack appearance under small loading only. For anode in battery application, the zero skewness is also required since this profile usually have the most larger contact area than other skewness. Large contact area can cause the lithium-ion easily connected to graphite, which means in macroscale, the battery has low electric resistance.

Another indicator commonly used in surface analysis is BAC curve. Figure 3b shows the BAC curves for each specimen. The original concept of BAC curve is to evaluate worn height after polishing treatment. However, it might be difficult to find reference surface point since in this work, these curves are obtained from different specimen that, in microscale, might not be identic. The BAC curves are plotted based on presumptions that by polishing the graphite with cellulose-based paper producing smoother surface, the deepest valley on the surface does not contact to the cellulose-based paper. As
consequence, two points from unpolished and cellulose-based paper are coincident in bearing length of 100%. In contrast, by polishing the graphite with P800, P400, and P220 sandpapers producing rougher surface, the highest peak does not contact to the sandpaper. Thus, the points in bearing length of 0% are coincident. Even though these presumptions are applied, the BAC curves still can give important information related to the surface characteristics.

As expected, it can be seen that by polishing the graphite specimen with cellulose-based paper, the graphite surface profiles become smoother, proven by the BAC curve of cellulose-based paper is flatter than that of the unpolished specimen. In contrast, polishing the graphite specimen with three different kinds of sandpapers yielded rougher graphite specimens, proven by the BAC curve of P800, P400, and P220 are slightly tilted compared to BAC curve of an unpolished specimen. Considering the prior and post polishing condition in BAC curves, the alteration of height variation at the surface can be plotted as shown in Figure 4. The height variation due to polishing treatment by cellulose-based paper can be reduced up to 45%. Furthermore, polishing the graphite surface by P800, P400, P220 can increase the height variation to 70%, 110%, and 220%, respectively.

The quantitative analysis of height variation can be used to determine which polishing treatment and its sandpaper suitable for design requirement. For example, surface contact area for mechanical locking in graphite-based composite or for electrical resistance in lithium-ion battery can be predicted based on its height variation. Typically, higher height variation indicates wider surface contact area. However, there is still remaining issue in analyzing surface characteristics. In fact, all of parameters investigated in this study still cannot determine the types of the surface profile. The best indicator that can be used for expecting a certain surface profile occur is based on ADF curve only. Some parameters that can directly determine surface profile needs to be introduced in the future. Thus, controlling surface region particularly in graphite can be conducted accurately.

![Figure 3](image1.png)

**Figure 3.** (a) ADF curves and (b) BAC curves of specimens with different polishing treatment.

![Figure 4](image2.png)

**Figure 4.** Height variation for each specimen.
4. Conclusion

The surface profile and roughness of graphite have been observed in this work. The influence of polishing treatment on the graphite surface is revealed. It can be seen that four parameters i.e. $R_a$, $R_q$, $R_p$, and $R_t$ show a similar tendency with surface roughness. Furthermore, the surface profile of each specimen is also analyzed by ADF and BAC curves. All specimens have nearly zero skewness tendency which means those can be useful for mechanical locking in case of designing graphite-based composite, and for reducing the electric resistance of an anode in lithium-ion battery application. Based on the analyses of surface parameters, it is still difficult to predict surface profile. Those parameters cannot differentiate surface profile prior polishing treatment i.e. continuous peaks and valleys and post polishing treatment i.e. scattered roughness. There is only ADF curve that can be indicator of surface profile. Otherwise, visual observation under microscope must be conducted to clearly capture the surface profile. Finding of novel parameter which able to indicate not only surface roughness but also its profile might be interesting for the future work.

Acknowledgments

This paper is funded by USAID through Sustainable Higher Education Research Alliances (SHERA) program with grant number IIE0000078-ITB-1.

References

[1] H Marsh and F R Reinoso 2006 Activated carbon. Elsevier.
[2] R G Sheppard, D M Mathes and D J Bray 2001 Properties and characteristics of graphite for industrial applications. Poco Graphite, pp. 5-7.
[3] Z Mirski 2011 Brazing a graphite composite to molybdenum alloy TZM using active copper-based filler metals with chromium additive. Archives of Metallurgy and Materials, 56(3), pp. 829-37.
[4] P I Zolkin 2005 Study of graphite materials to be used in medicine. International Polymer Science and Technology, 32(9), pp. 67-8.
[5] S K Choudhury and S Chinchankar 2016 1.3 Finish Machining of Hardened Steel. Comprehensive Materials Finishing, 29, pp. 47.
[6] E S Gadelmawla, M M Koura, T M Maksoud, I M Elewa and H H Soliman 2002 Roughness parameters. Journal of Materials processing Technology, 123(1), pp. 133-45.
[7] B A Budiman, F B Juangsa, M Aziz, I P Nurprasieto and I N Zaini 2017 Experimental Verification of Interfacial Strength Effect on the Mechanical Properties of Carbon Fiber-Epoxy Composite. International Journal on Advanced Science, Engineering and Information Technology, 7(6), pp. 2226-31.
[8] B A Budiman, F Adziman, P L Sambegoro, I P Nurprasieto, R Ilhamsyah and M Aziz 2018 The Role of Interfacial Rigidity to Crack Propagation Path in Fiber Reinforced Polymer Composite. Fibers and Polymers, 19(9), pp. 1980-8.
[9] M Yoshio, H Wang, K Fukuda, T Umema, T Abe and Z Ogumi 2004 Improvement of natural graphite as a lithium-ion battery anode material, from raw flake to carbon-coated sphere. Journal of Materials Chemistry, 14(11), pp. 1754-8.
[10] M. Mundzinger, S Farsi, M Rapp, U Golla-Schindler, U Kaiser and M Wachtler 2017 Morphology and texture of spheroidized natural and synthetic graphites. Carbon, 111, pp. 764-73.
[11] T Zhang, J Gao, L J Fu, L C Yang, Y P Wu and H Q Wu 2007 Natural graphite coated by Si nanoparticles as anode materials for lithium ion batteries. Journal of Materials Chemistry, 17(13), pp. 1321-5.
[12] P Petrik, L P Biró, M Fried, T Lohner, R Berger, C Schneider, J Gyulai and H Ryssel 1998 Comparative study of surface roughness measured on polysilicon using spectroscopic
ellipsometry and atomic force microscopy. *Thin Solid Films*, **315**(1-2), pp. 186-91.

[13] K Y Lee, M C Kang, Y H Jeong, D W Lee, J S Kim 2001 Simulation of surface roughness and profile in high-speed end milling. *Journal of Materials Processing Technology*, **113**(1-3), pp. 410-5.

[14] O B Abouelatta and J Madl 2001 Surface roughness prediction based on cutting parameters and tool vibrations in turning operations. *Journal of materials processing technology*, **118**(1-3), 269-77.