

## THE POTENTIAL OF INDONESIAN GRAPHITE AS RDE FUEL MATRIX

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### ABSTRACT

**THE POTENTIAL OF INDONESIAN GRAPHITE AS RDE FUEL MATRIX.** The development plan of Experimental Power Reactor (RDE) in Indonesia is non-commercial and leads to the technology type of Pebble Bed Reactor (PBR) - High Temperature Gas Cooled Reactor (HTGR). The fuel used for PBR reactors is kernel dispersed in spherical fuel elements. The matrix used in PBR nuclear fuel is graphite which functions as a neutron moderator, fuel protective material and heat conductor. Domestication of the domestic fuel matrix needs to be conducted to improve national independence. Therefore, it is necessary to do research on the potential of local graphite to be used as RDE fuel matrix. This study focused on the identification and characterization of local and commercial graphite. The results are compared with the literature, how far it is fulfilling nuclear grade graphite for PBR fuel matrix. Characterization of graphite includes phase analysis with XRD, microstructure with SEM, surface area/porosity, impurities determination with AAS, ICP-OES and NAA, equivalent boron content, carbon content, density, particle size distribution and ash content. The characterization results show that the carbon content obtained was  $87.0 \pm 4.2\%$  for local graphite and 100% for commercial graphite. Meanwhile, for the purposes of nuclear graphite it requires a carbon content of  $>99\%$ . The impurity content in local and commercial graphite still does not meet the RDE fuel matrix standard. The results of XRD analysis show that the local graphite phase is the same as the commercial graphite phase, namely the 2H graphite hexagonal crystal system with the lattice group of P 63/mmc. Particle size distribution and surface area of local graphite are higher compared to nuclear graphite literature. The ash content of commercial graphite was  $0.236 \pm 0.029$  and local graphite was  $9.587 \pm 0.010\%$ . The results of this study indicate that the local graphite from the flotation still requires a further refinement process to obtain local graphite that can be used as a fuel matrix for RDE.

**Keywords:** Natural graphite, matrix, fuel, PBR, RDE, characterization

### ABSTRAK

**POTENSI GRAFIT INDONESIA SEBAGAI MATRIKS BAHAN BAKAR RDE.** Rencana pembangunan Reaktor Daya Eksperimental (RDE) di Indonesia bersifat non-komersial dan mengarah ke teknologi jenis *Pebble Bed Reactor* (PBR) – *High Temperature Gas Cooled Reactor* (HTGR). Bahan bakar yang digunakan untuk reaktor PBR adalah kernel yang terdispersi dalam elemen bakar sferis (bola). Matriks yang digunakan pada bahan bakar nuklir PBR adalah grafit yang berfungsi sebagai moderator neutron, material pelindung bahan bakar dan konduktor panas. Domestifikasi matriks bahan bakar dalam negeri perlu dilakukan untuk meningkatkan kemandirian bangsa. Oleh karena itu, perlu dilakukan penelitian tentang potensi grafit lokal untuk digunakan sebagai matriks bahan bakar RDE. Penelitian ini difokuskan pada identifikasi dan karakterisasi grafit lokal dan komersial. Hasilnya dibandingkan terhadap literatur, seberapa jauh memenuhi grafit berderajat nuklir untuk matriks bahan bakar PBR. Karakterisasi grafit meliputi analisis fasa dengan XRD, struktur mikro dengan SEM, luas permukaan/porositas, penentuan impuritas dengan AAS, ICP-OES and NAA, boron ekuivalen, kandungan karbon, densitas, distribusi ukuran partikel dan kadar abu. Hasil karakterisasi menunjukkan bahwa kandungan karbon diperoleh  $87,0 \pm 4,2\%$  untuk grafit lokal dan 100% untuk grafit komersial. Sementara, untuk keperluan grafit nuklir membutuhkan kandungan karbon  $>99\%$ . Kandungan impuritas dalam grafit lokal dan komersial masih belum memenuhi standar matriks bahan bakar RDE. Hasil analisis XRD menunjukkan bahwa fasa grafit lokal sama dengan fasa grafit komersial yaitu grafit 2H sistem kristal heksagonal dengan grup kisinya adalah P 63 /mmc. Distribusi ukuran partikel dan luas permukaan grafit lokal lebih tinggi dibanding dengan grafit nuklir literatur. Kadar abu grafit komersial diperoleh  $0,236 \pm 0,029$  dan grafit lokal sebesar  $9,587 \pm 0,010\%$ . Hasil dari penelitian ini, menunjukkan bahwa grafit lokal hasil flotasi masih memerlukan proses pemurnian lebih lanjut supaya diperoleh grafit lokal untuk dapat digunakan sebagai matriks bahan bakar untuk RDE.

**Kata kunci:** Grafit alam, Matriks, Bahan bakar, PBR, RDE, karakterisasi

## INTRODUCTION

Construction of non-commercial Experimental Power Reactors (RDE) leads to the technology type of Pebble Bed Reactor (PBR) - High Temperature Gas Cooled Reactor (HTGR). This type of reactor was chosen as it has very high safety features due to its passive safety features in the form of the ability to manage cooling performance without the need for external assistance and flexible applications, besides being able to generate electricity, it also produces high temperature steam which can be used for coal liquefaction, water desalination and water production [1]. The fuel used for PBR reactors is a kernel dispersed in spherical fuel elements. Kernel fuel contains  $\text{UO}_2$  called TRISO (Tris-structural-isotropic) particles coated with buffers, Inner Pyrocarbon (Ipyc), SiC and Outer Pyrocarbon (Opyc). These kernel fuels are in the thousands, dispersed in the graphite matrix of fuel elements [2,3,4].

PBR fuel matrix is an A3-3 graphite matrix with a composition of natural graphite of 64%, synthetic graphite 16% and phenolic resin 20%. With the mixture of natural graphite and synthetic graphite, the material obtained can be compressed; the material is quite strong and purer. This characteristic is obtained from the nature of synthetic graphite which has high purity and is less anisotropic and natural graphite which can be reduced in size to very fine [2,5,6].

Efforts for the RDE program can continue by leading to Nuclear Power Plants (NPP) and to create energy sovereignty, then Indonesia needs to conduct Development on RDE Fuel Fabrication Technology. Mastery of RDE fuel fabrication technology for power reactor pilot projects with a capacity of around 10 MW with the HTR-10 Pebble Bed Reactor (PBR) type, including the mastery of kernel production technology, production of TRISO coated particles, and fuel spheres to produce fuel that meets required specifications. In addition to the need for nuclear-grade fissile material that can split in the reactor, moderator materials and the structure of the fuel protective material in accordance with the requirements are also necessary. The graphite matrix functions as a neutron moderator, fuel protective material structure, and heat transfer media [7].

Graphite is one of the allotropes of crystalline carbon which has good nuclear physical characteristics such as high efficiency of reflection and moderation, relatively low atomic mass, low neutron uptake latitude, high mechanical strength, high temperature resistance, easy fabrication and light mass [5]. Natural graphite as a constituent of A3 – 3 graphite matrix used in PBR type fuels must meet the criteria of physical and chemical properties including purity, impurity content, boron equivalent content, surface area, particle size distribution, ash content and density. Graphite is a natural element with a carbon composition (C) with a hexagonal crystal system and has a density of  $2.23 \text{ g/cm}^3$ . Yeo et. al, Tang et. al and Zhao et. al use natural graphite as HTGR fuel matrix with high purity ( $> 99\%$ ), lithium content  $< 0.005 \text{ } \mu\text{g/g}$ , boron equivalent content  $< 1 \text{ } \mu\text{g/g}$

and ash content  $\sim 100 \text{ } \mu\text{g/g}$  with graphite surface area used of  $4.8 - 5.5 \text{ m}^2/\text{g}$ , with microstructure form which is more flattened and sharper compared to synthetic graphite. Graphite particle size distribution used by Yeo et. al and Tang et. al is 90% with a range of  $32 - 63 \text{ } \mu\text{m}$ , Zhao et. al used graphite with a size of  $< 80 \text{ } \mu\text{m}$  with 71.20% of  $< 25 \text{ } \mu\text{m}$ . The particle size distribution based on TecDoc 1674 is 50% particle size of  $< 32 \text{ } \mu\text{m}$  [2,3,4].

In China, around 60 tons of graphite has been used for fuel of High Temperature Gas - Cooled Reactor (HTR) - 10 and more than 1000 tons will be used for High Temperature Gas - Cooled Reactor Pebble-Bed Module (HTR-PM) [8]. It takes around 27000 PBR fuel for initial operation of RDE and 105 fuel per day to achieve steady state [1]. The need for graphite matrix per fuel is 0.2 kg so it takes about 156 tons of graphite for 20 years to operate 1 RDE with a capacity of 10 MW.

Graphite research for the nuclear sector began in 2007 in the form of synthetic graphite derived from calcine coke and Tar Pitch [9]. Natural graphite from Sulawesi has been refined by flotation method and obtained a carbon content of 61% [10]. Natural graphite from Kalimantan has been purified by flotation and acid process (HF) methods and obtained a carbon content of more than 90% [11,12]. Flotation method is a method of separation and concentration based on differences in inter-phase chemical properties. Graphite naturally floats in water because it is hydrophobic; some suitable reagents are added to increase its recovery [10].

From the research, the results of the study have not been obtained on the potential of Indonesian natural graphite to be used as RDE matrix with higher purity and characteristics according to specifications. To domesticate the domestic fuel matrix and improve national independence, it is necessary to study local graphite as an RDE fuel matrix. This study aims to figure out the potential of local graphite as RDE fuel matrix and commercial (synthetic) graphite as a comparison.

Indonesia has quite a lot of graphite mineral deposits in Sulawesi and Kalimantan regions. In Sulawesi, graphite rocks are found in Tamboli, Samaturu Subdistrict, Kolaka Regency, Southeast Sulawesi with 3% graphite content and the main compound was calcite ( $\text{CaCO}_3$ ) and explored by PT Mekongga Sejahtera [10] with an exploration permit covering 98 ha. In Sulawesi, graphite is also found in Latambaga Subdistrict, Kolaka Regency, Southeast Sulawesi in the form of slate with the result of major chemical graphite minerals is  $\text{SiO}_2$  with a percentage of 29.08% -70.76% and carbon content of 2.36% - 4.51% [13].

In Kalimantan, graphite is found in Balai Sebut Subdistrict, Sanggau Regency, West Kalimantan identified as having graphite mineral content along with other minerals with the main compound of  $\text{SiO}_2$  24.27 - 90.12%, with carbon content of 1.25% - 65.56%

and mineral in the form of slate mud stone [13]. In Sanggau Regency, PT. Trans Sulawesi Sejahtera holds a graphite mining business permit with an area of 10,000 Ha and PT. Trans Sulawesi Tenggara (TST) with an area of 80.5 Ha.

In Sumatra, graphite is found in Muara Saiti Pangkalan and Tanjung Balit, Lima Puluh Kota Regency (an insertion in schist rocks), in Ombilin in the Singkarak Lake area and in Siberlabu Payakumbuh [14].

## EXPERIMENTAL METHOD

### Materials and Equipment

The materials used were natural graphite from Kalimantan, HNO<sub>3</sub> pro analysis (Merck), H<sub>2</sub>SO<sub>4</sub> pro analysis (Merck), Element Standards AAS and ICP - OES (Merck), HP acetylene gas, HP nitrous oxide gas, UHP argon gas, and UHP helium gas.

The equipment used was XRD (Panalytical) for crystallography, particle size analyzer (PSA) micro with the brand of Cilas 1190 Liquid with the Fraunhofer method for particle size distribution, Scanning Electron Microscopy with Energy Dispersive Spectroscopy (SEM-EDS) Jeol 6510LA and Carbon and Sulfur Analyzer, (CS 744 LECO) for carbon content. He - Ultrapyc 1200-e autopycnometer, Quantachrome for determination of density, Bicasa-brand Furnace for determination of ash content. Neutron Activation Analysis (NAA), flame - Atomic absorption Spectrometer (F-AAS) and Agilent-brand ICP - OES for determination of impurity and boron equivalent and high performance microwave digester systems area used for preparation. Surface area and pore size analyzer Quadrasorb SI, Quantachrome is for determining the surface area of Brunauer Emmet Teller (BET) and porosity. Scanning Electron Microscopy (SEM) Jeol 6510LA is for microstructure analysis.

### Procedure

Chemical and physical characterization was carried out to figure out the potential and source of graphite in Indonesia as RDE fuel matrix. Natural graphite from Kalimantan was initially refined using the flotation method [11], then identified and characterized, and the results were compared with commercial (synthetic) graphite from Merck. Characterization includes crystallography, particle size distribution, carbon content, density, ash content, impurity and equivalent boron content, BET surface area, porosity and microstructure.

Crystallographic characterization was performed using XRD (Panalytical), particle size distribution used micro particle size analyzer (PSA) Cilas 1190 Liquid with the Fraunhofer method, the carbon content was determined by SEM/EDS Jeol 6510LA using ZAF method standardless quantitative analysis and Carbon and Sulfur Analyzer, (CS 744 LECO). Determination of density used He - autopycnometer Ultrapyc 1200-e, Quantachrome. Ash content was determined by the loss

on ignition method at a temperature of  $\pm 950^{\circ}\text{C}$ . Impurities and boron equivalent used Neutron Activation Analysis (AAN), Flame - Atomic Absorption Spectrometer (F-AAS) and Agilent-brand ICP - OES. Measurements with AAN, samples were analyzed without treatment while for AAS and ICP-OES, graphite samples were dissolved first with a mixture of acids (HNO<sub>3</sub> and H<sub>2</sub>SO<sub>4</sub>) in a microwave digester. BET surface area and porosity were determined using Surface area and pore size analyzer Quadrasorb SI, Quantachrome. Graphite SEM images were taken using Scanning Electron Microscopy (SEM) Jeol 6510LA.

## RESULTS AND DISCUSSION

### Characterization of commercial graphite and Indonesian (local) natural graphite

The results of the analysis of crystallographic diffraction patterns of local and commercial graphites with 2 Theta 20 - 80 are presented in Figure 1. The results of diffractogram comparison of commercial graphite shown in Figure 1 were performed using Highscore Plus application from Panalytical using COD database. The results show that there is a phase match for commercial graphite with card numbers 96-101-1061. The compatibility of the graphite phase obtained on card numbers 96-101-1061 is 2H graphite with a hexagonal crystal system and the lattice group is P 63 m c with a peak of 2  $\Theta$  26.228°, 42.214°, 44.365°, 50.381°, 53.974°, 59.404°, 70.804°, 77.177°. Based on the semi quantitative calculation results, the percentage of 2H graphite in commercial graphite (red diffractogram) is 100 percent, while for natural graphite (blue diffractogram), there is a phase similar to commercial graphite, but there is a peak residual at 2  $\Theta$  28.442°, 36.10°, 40°, 43.94° and 73.889° which indicate that natural graphite contains another large phase. The results of phase compatibility of the COD database against the residual peaks obtained include copper with cards number 96-901-3016, with a cubic crystal system and lattice group of Fm-3m with a peak at 2 $\Theta$  43.94°; 50.21° and 74.15°. Rietveld method is used to determine the c/a ratio and is obtained for commercial graphite and natural graphite of 2.7411 and 2.7294, respectively. The c/a ratio obtained is lower compared to 2H graphite with cards number 96-101-1061, as 2.7489.

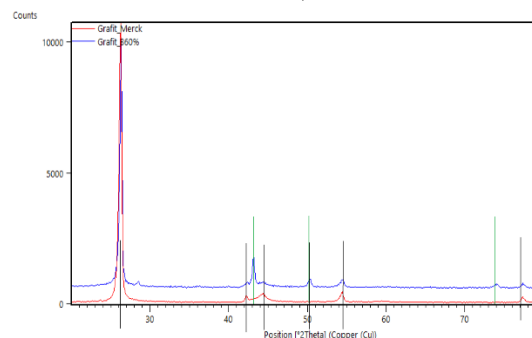


Figure 1. X-ray diffraction pattern of commercial and local graphite. Black line is 2H graphite, green line is Cu metal.

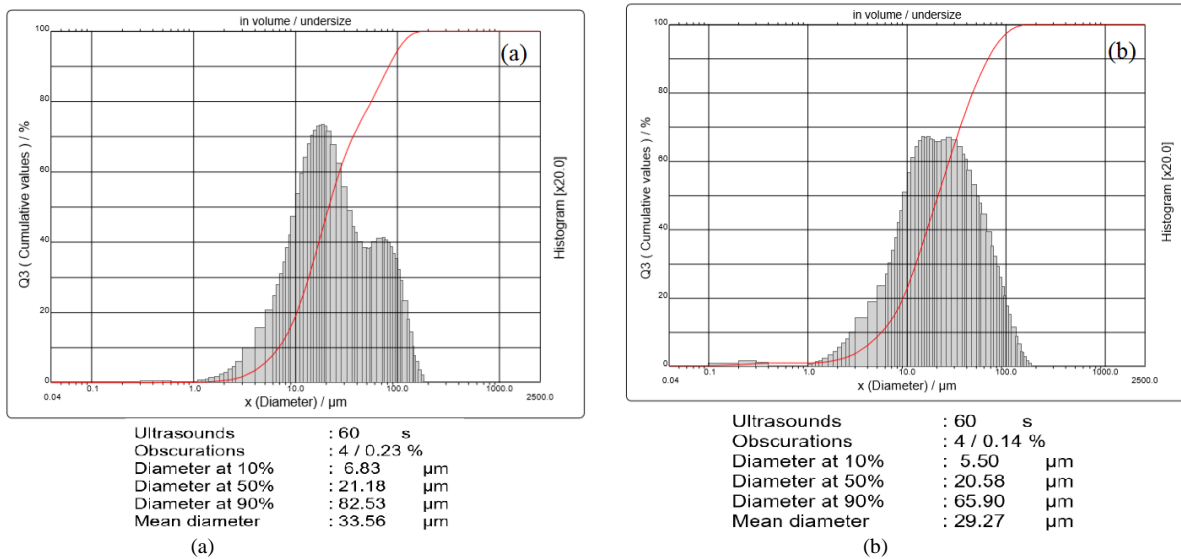


Figure 2. Particle size distribution of (a) commercial graphite and (b) local graphite

The results of the density measurement of commercial graphite is obtained at 2.311 g/cm<sup>3</sup> and local graphite at 2.334g/cm<sup>3</sup>. Graphite density obtained both commercially and locally is higher than the graphite density in the literature (graphite density value in literature is 2.09 - 2.266 g/cm<sup>3</sup>) [13,14]. Zhao et al explained that the density of graphite as a spherical nuclear fuel matrix is greater than 1.70 g/cm<sup>3</sup> [3].

The results of particle size distribution were determined using particle size analyzer Cilas as shown in Figure 2. Figure 2(a) shows that commercial graphite has an average particle size of 33.56 μm with a distribution of 10% to 90% is 6.83 - 82.53 μm, while local graphite as shown in Figure 2(b) has a average particle size of 29.27 μm with a distribution of 10% to 90% is 5.50 - 65.90 μm. These results indicate that commercial graphite has a larger particle size distribution than local graphite. Meanwhile for the requirements for nuclear fuel needs, the dominant size of graphite diameter is < 25 μm [3] and 32 - 63 μm [4]. Based on this particle size distribution, both local graphite and commercial graphite have a greater range of requirements as nuclear graphite. The large average diameter and particle size distribution have an effect on the fabrication process and the graphite matrix density produced after compacting. Larger powder diameters cause many cavities so that smaller powder is needed to fill the cavity (as a filler). If the cavity is not filled, the density of the graphite ball will be lower and the graphite matrix will be easily cracked (low mechanical strength) [3].

The spectrum of measurement of carbon content with EDS is shown in Figure 3. The measurement results obtain the percentage of carbon mass of commercial graphite (Figure 3(a) as 100 ± 0.16% and local graphite (Figure 3(b) 89.68 ± 0.06% with the fitting coefficient is 0.3999 and 0.1994, re-spectively. These results are almost the same as the determination of carbon using a carbon analyzer. The amount of carbon content in commercial graphite is obtained as 100 ±

1.3% and local graphite as 87.0 ± 4.2%. The carbon content of commercial graphite has met the requirements as nuclear graphite (nuclear graphite carbon content > 99%) [3], however, local graphite has not met them, so it still requires further processing to refine the carbon content to reach > 99%.

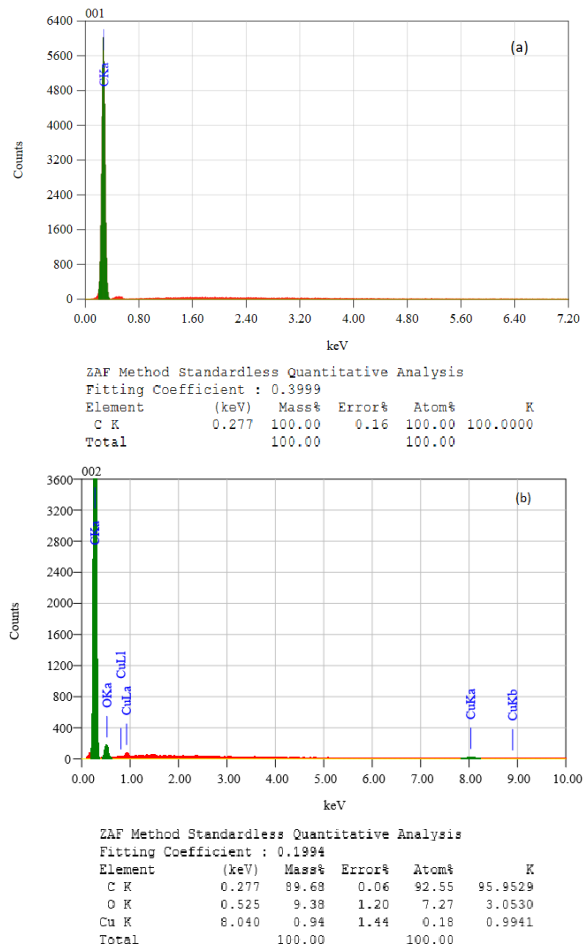


Figure 3. EDS Spectrum of (a) Commercial graphite and (b) Local graphite

Determination of graphite impurity is conducted by several methods; so that the results of the overall elemental impurity analysis in graphite can be known as each measurement method has different analytical skills. There are two analytical methods for determining impurity in graphite, namely; non-destructive test with neutron activation analysis (NAA) and destructive test with AAS and ICP-OES. Table 1 shows the results of impurity determination in local and commercial graphites with AAS, AAN and ICP-OES analysis methods. Table 1 shows that there are 31 elements determined by the three methods. In general, impurity concentrations in local graphite are higher compared to commercial graphite. In local graphite, there are 18 elements that can be determined, namely; Al, Co, Cr, Cu, Dy, Fe, K, La, Mg, Mn, Na, Ni, Pb, Sb, Sc, V, W and Zn, whereas other elements such as Ag, B, Ba, Ca, Ce, Cd, Eu, Hf, Li, Si, Sm, Th and Ti are not detected/below the limit of detector. The highest concentration of elements obtained in Cu reached 45660 µg/g (4.5%), followed by Dy 24540 µg/g (2.45%), Sb 2324 µg/g (0.232%), Fe 1379 µg/g (0.1379%) and Al 1071 µg/g (0.1071%). Elements of Mg, Pb, and Zn are in the concentration range of 190 to 451 µg/g and concentration of others is below 100 µg/g. Whereas for commercial graphite, it is known that 24 elements, with the highest element content is obtained for Dy element at 6805,833 µg/g followed by Fe 433 µg/g and Cr 144 µg/g, and others below 100 µg/g.

The presence of impurity in graphite results in a catalytic effect on carbon elements. The catalytic effect of this metal can accelerate the oxidation of graphite structures [15], and cause fuel failure due to the chemical reaction between carbon and impurity [16].

The magnitude of the effect of the oxidation speed depends on the type and concentration of impurity. The presence of impurity at a certain level can increase the speed of graphite oxidation to 2-3 times. There are several elements that have a catalytic effect on graphite, namely; Ti, V, Cr, Fe, Mo, W, Re, Ru, Rh, Ir, Cu, Zn, Pd, Ag, Pt and Eu. At nuclear graphite with high purity, total impurity concentration does not exceed 300 µg/gr. As graphite with low purity, the impurity concentration is allowed to be a maximum of 1,000 µg/g [17]. High impurity content can also reduce resistance to corrosion, reduce thermal conductivity and reduce irradiation performance of fuel [3].

Table 1. Impurity data of commercial and local graphites

Element	Content (µg/g)		Method
	Commercial graphite	Local graphite	
Ag	49.740±0.0990	< LoD 0.2823	AAS
Al	39.59 ± 5.25	1071.35 ± 40.60	AAN
B	<LoD 12.501	< LoD 12.5012	AAS
Ba	10.46 ± 1.80	Not detected	AAN
Ca	<LoD 0.3819	< LoD 0.3819	AAS
Ce	1.44 ± 0.04	Not detected	AAN
Cd	< LoD 0.0015	< LoD 0.0015	AAS

Co	0.40 ± 0.01	7.98 ± 0.83	AAN
Cr	144.027 ± 0.1798	74.800 ± 0.140	AAS
Cu	65.322 ± 0.160	45660 ± 72.00	AAS
Dy	6805.833	24540	ICP
Eu	0.02 ± 0.001	Not detected	AAN
Fe	433.080 ± 0.999	1379.20 ± 80.00	AAS
Hf	0.03 ± 0.001	Not detected	AAN
K	8.640 ± 0.02	78.080 ± 0.620	AAS
La	0.73 ± 0.04	1.05 ± 0.09	AAN
Li	< LoD 1.8600	< LoD 1.8600	AAS
Mg	6.992 ± 0.998	253.400 ± 4.60	AAS
Mn	5.31 ± 0.31	10.33 ± 0.73	AAN
Na	84.90 ± 1.56	85.10 ± 1.60	AAN
Ni	60.927 ± 0.1797	70.600 ± 0.24	AAS
Pb	< LoD 60.500	190 ± 0.18	AAS
Sb	0.40 ± 0.01	2324.59 ± 273.10	AAN
Sc	0.25 ± 0.01	0.44 ± 0.03	AAN
Si	< LoD 1,2000	< LoD 1,2000	AAS
Sm	0.03 ± 0.001	Not Detected	AAN
Th	0.16 ± 0.01	Not Detected	AAN
Ti	< LoD 1.200	< LoD 1.200	AAS
V	3.61 ± 0.39	34.04 ± 1.33	AAN
W	0.38 ± 0.03	15.00 ± 0.86	AAN
Zn	3.77 ± 0.24	451.27 ± 31.73	AAN

Other types of impurity that need to be avoided in the graphite matrix are the elements that contribute to absorbing neutrons. The neutron absorption rate of an element of impurity can be expressed by Equivalent Boron Content-EBC as shown in equation 1 [18].

$$\text{Total EBC} = \sum \left( \frac{\text{EBC Factors}}{\text{impurity content } (\mu\text{g/g})} \right) \dots \dots \dots (1)$$

In determining EBC, it is known that elements B, Dy, Eu, Gd, Sm, Li and Cd are impurities with a large EBC factor, each of which is 1.000; 0.0818; 0.4250; 4.3991; 0.5336; 0.4139 and 0.3172. Addition to the 6 elements above, the element EBC factor is 0.0001 to 0.0263 with a small contribution to neutron absorption. In Table 1, the content of elements B, Eu, Gd, Sm, Li and Cd is not detected, while the Dy concentration in commercial graphite is relatively high (6805.85 µg/g) and local graphite as 24540 µg/g. This equivalent boron content is higher than nuclear graphite at < 1 µg/g [3] and < 3.0 µg/g [2].

Images of commercial graphite and local graphite are presented in Figure 4. Commercial graphite morphology looks flatter with sharper sides compared to local graphite. The morphological form is similar to graphite powder produced by Sunghwan Yeo, et.al. [4].

The surface area of commercial and local graphite was obtained respectively at 10.54 and 11.41 m<sup>2</sup>/g. In some literature it is stated that the graphite surface area with nuclear grade varies in the range of 1.15 m<sup>2</sup>/g to 5.55 m<sup>2</sup>/g [3,4], however, the surface area obtained in both local graphite and commercial graphite is still higher compared to H graphite from Zhao, et. al and S. Yeo, et.al. This higher surface area might be re-



lated to graphite particle size. Pore volume of local graphite is higher than commercial graphite. This is related to the impurity content in the graphite structure. With the presence of particle impurity which is sufficiently high in the graphite structure, it forms cavities between the graphite structures so that the local graphite pore volume is higher.

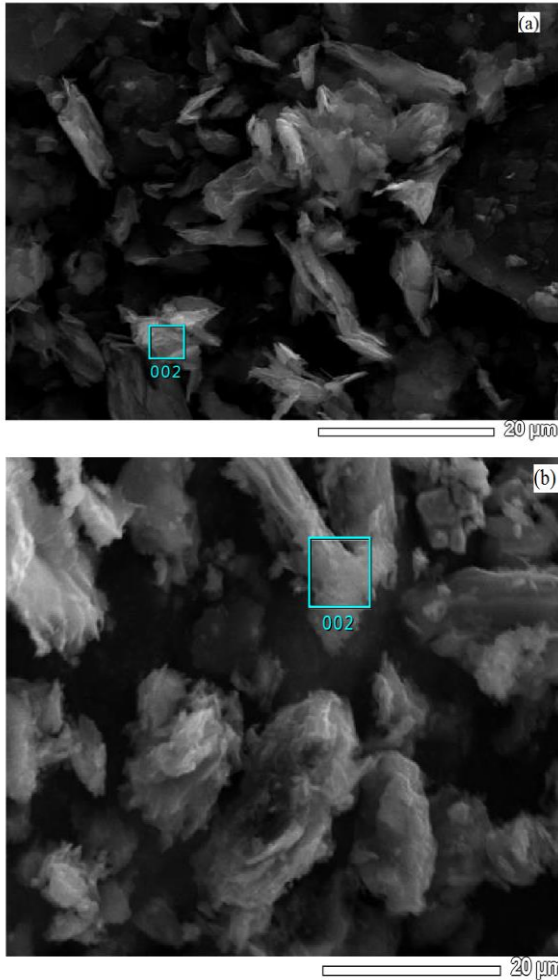


Figure 4. SEM images of (a) Commercial Graphite (b) Local Graphite.

BET surface area and porosity are closely related to the corrosion properties of graphite. The greater the surface area, the resistance to corrosion decreases. High porosity will affect electrical resistance, heat conductivity, elastic modulus, material permeability and transport properties of fission products [19].

Pore size distribution is calculated using Density Functional Theory (DFT) method as can be seen in Figure 6. Local and commercial graphite has a similar pore structure. There are at least 3 pore size quantities observed in Figure 6, namely; pore sizes of 4 nm (most dominant), 7.5 nm and 14 nm. Clearly, the size of this pore diameter is in the range of 2 nm-50 nm, meaning that both graphites have a mesopore system.

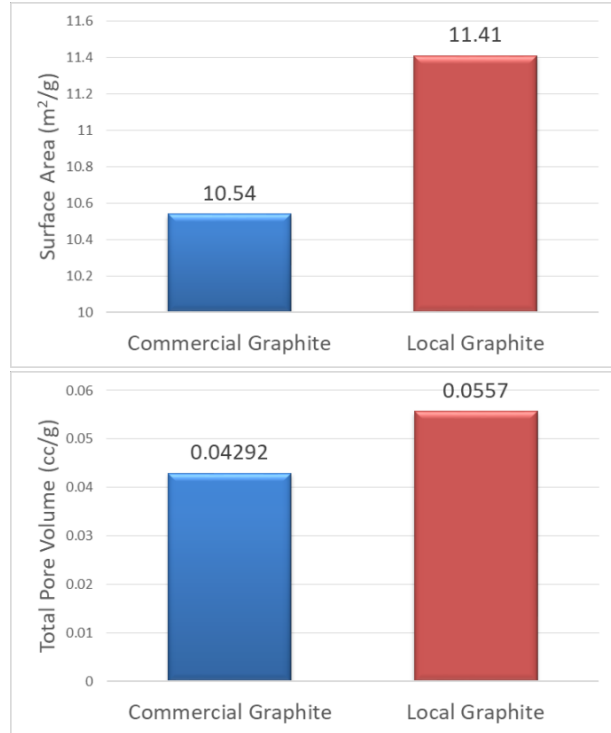


Figure 5. Surface area and total pore volume of commercial and local graphites.

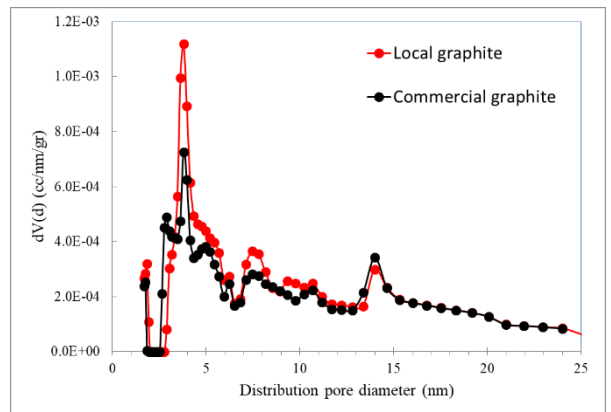


Figure 6. Pore size distribution with DFT method of Commercial Graphite and Local Graphite

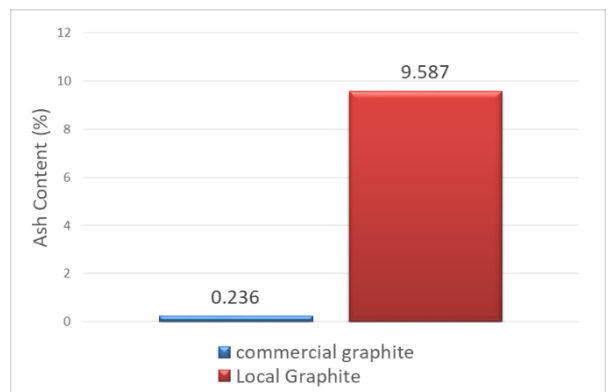


Figure 7. Ash contents of commercial and local graphites

The results of the ash content measurement of commercial graphite and local graphite are shown in Figure 7. Ash content of local graphite ( $9.587 \pm 0.010\%$ ) is greater than the ash content of commercial graphite ( $0.236 \pm 0.029\%$ ). Sunghwan Yeo et.al. mention nuclear graphite ash content  $< 100$  ppm [4]. The results show that the impurity content in commercial graphite and local graphite is still quite high.

## CONCLUSION

Indonesian graphite has the potential to be refined into nuclear graphite to meet the construction needs of Experimental Power Reactor. However, the results of various characterizations show that natural graphite which has been treated by flotation still does not meet the criteria for nuclear graphite, especially in its impurity content. The carbon content of graphite from flotation is 87.0%, meaning that there are still around 13% of materials other than carbon in graphite. These data are supported by the results of determining the content of metals in graphite which is still quite high. Continuing this research, graphite from the flotation still requires further processing, especially in terms of refinement. This can be conducted by chemical or thermal means to eliminate impurity so that it meets the criteria as nuclear graphite to meet the material needs of RDE fuel matrix.

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## REFERENCES

- [1]. T. Setiadipura, S. Bakhri, G. R. Sunaryo, and D. S. Wisnubroto, "Cooling passive safety features of Reaktor Daya Eksperimental," *AIP Conf. Proc.*, vol. 1984, 2018.
- [2]. C. Tang, Y. Tang, J. Zhu, Y. Zou, J. Li, and X. Ni, "Design and manufacture of the fuel element for the 10 MW high temperature gas-cooled reactor," *Nucl. Eng. Des.*, vol. 218, no. 1–3, pp. 91–102, 2002.
- [3]. H. Zhao, T. Liang, J. Zhang, J. He, Y. Zou, and C. Tang, "Manufacture and characteristics of spherical fuel elements for the HTR-10," *Nucl. Eng. Des.*, vol. 236, no. 5–6, pp. 643–647, 2006.
- [4]. S. Yeo, J. Yun, S. Kim, M. S. Cho, and Y. W. Lee, "Fabrication methods and anisotropic properties of graphite matrix compacts for use in HTGR," *J. Nucl. Mater.*, vol. 499, pp. 383–393, 2018.
- [5]. IAEA, "High Temperature Gas Cooled Reactor Fuels and Materials," *IAEA Tecdoc 1645*, pp. 1–182, 2010.
- [6]. P. P. Magampa, N. Manyala, and W. W. Focke, "Properties of graphite composites based on natural and synthetic graphite powders and a phenolic novolac binder," *J. Nucl. Mater.*, vol. 436, no. 1–3, pp. 76–83, 2013.
- [7]. X. Zhou et al., "Study on the Comprehensive Properties and Microstructures of A3-3 Matrix Graphite Related to the High Temperature Purification Treatment," *Hindawi Sci. Technol. Nucl. Install.*, 2018.
- [8]. X. W. Zhou, Y. P. Tang, Z. M. Lu, J. Zhang, and B. Liu, "Nuclear graphite for high temperature gas-cooled reactors," *Xinxing Tan Cailiao/New Carbon Mater.*, vol. 32, no. 3, pp. 193–204, 2017.
- [9]. T. Indrayati and Sudaryadi, "Penyiapan umpan grafitisasi dengan metoda granulasi bertahap," *Pros. PPI - PDIPTN*, pp. 278–282, 2007.
- [10]. F. F. Florena et al., "Floatability study of graphite ore from southeast Sulawesi (Indonesia)," *AIP Conf. Proc.*, vol. 1712, 2016.
- [11]. I. M. Joni et al., "Synthesis and dispersion of nanoparticles, and Indonesian graphite processing," *AIP Conf. Proc.*, vol. 1554, pp. 20–26, 2013.
- [12]. C. Panatarani, A. O. Maulana, A. Rianto, and I. M. Joni, "Preparation of graphite oxide by sodium cholate intercalation and sonication from Indonesian natural graphite," *AIP Conf. Proc.*, vol. 1712, no. March 2016, 2016.
- [13]. R. Marza, W. Setiyawan, and H. Rodiana, "Ke prospekan Mineral Grafit di Pulau Kalimantan dan Pulau Sulawesi," *Pros. Has. Kegiat. Pus. Sumber Daya Miner. Batubara dan Panas Bumi Tahun Anggar. 2017*, vol. Buku 2 : B, pp. 219–229, 2018.
- [14]. B. J. Marsden, G. N. Hall, 2012, "Graphite in gas - cooled reactors", *Comprehensive Nuclear Materials Volume 4*, Elsevier, Amsterdam, pp 325-390
- [15]. H.G. Maahs, D.R. Shryer, "Chemical impurity data on selected artificial graphites with comments on the catalytic effect of impurities on oxidation rate", *NASA Technical Note (NASA TN D-4212, National Aeronautic and Space Administration*, Washington, D.C. 1967.
- [16]. X. Luo, X. Wang, L. Shi, X. Yu, and S. Yu, "Nuclear graphite wear properties and estimation of graphite dust production in HTR-10," *Nucl. Eng. Des.*, vol. 315, pp. 35–41, 2017.
- [17]. W. Windes, G. Strydom, R. Smith, J. Kane, 2014, "Role of nuclear grade graphite in controlling oxidation in modular HTGRs", *IN/EXT-14-31720*, Idaho National Laboratory, U.S. Department of Energy National Laboratory. 2014.
- [18]. ASTM, "Standard Practice for Determining Equivalent Boron Contents of Nuclear materials C 1233". *ASTM International*. 1993

[19]. IAEA, “Advances in High Temperature Gas Cooled Reactor Fuel Technology,” in *IAEA*

*TECDOC CD 1674*, 2012, pp. 1–690.



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