

Preliminary Study of Rare Earth Element and Yttrium (REY) Content of Coal In Sangatta Coalfield, East Kalimantan, Indonesia

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Abstract - Kutai Basin is known as one of the most prolific sedimentary coal basins in Indonesia. Coal-bearing sequence in Kutai Basin is mainly Miocene to Eocene in age in which the coal seams are mostly in Miocene strata. Researchers have found that REY concentration in some coals and coal ashes are equal to or higher than that of the conventional deposit, as such coal deposit has become an important source for REY. Moreover, REY recovery as by-product from coal deposits could alleviate the "dirty" into "clean" coal energy. Referring to this shifted paradigm, a study on REY in Indonesian coal has put interest not only for researchers, but also for all stakeholders in this country. In this particular study, Sangatta coalfield in East Kalimantan was chosen as the area to observe the REY concentration. This area has been set due to its interesting geological setting, especially Pinang Dome that might have coal deposit with enriched REY. For the analyzed samples, drill cores were collected and then observed closely using polished section, inductively coupled plasma atomic emission spectroscopy (ICP-AES), and inductively coupled plasma mass spectrometry (ICP-MS). Based on the collected data, the highest REY concentration in coal deposit is located in the nearest part of the Pinang Dome. The REY content in coal deposit is associated with hydrothermal fluids and sediment source. These data suggest that there is a significant effect on Pinang Dome in the REY content in the coal deposit.

Keywords: rare earth element, yttrium, Sangatta, Kutai, coalfield

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INTRODUCTION

Rare earth elements and yttrium (REY) are fundamental commodities for several high technology industries and wide applications (Pecht *et al.*, 2012). The demand for REY increases significantly as high as 5.3% per year (Alonso *et al.*, 2012). The term of REY is used to define the elements including REE and yttrium. Due to expanding demand on REY and limited supply of conventional deposits (*e.g.* carbonatite, alkaline granites, and weathering crusts), some explorations and prospecting on "unconventional" deposits have been conducted. Goldschmidt and Peters (1933) have conducted the first study about REY in coal. Coal deposits become important alternative source for REY, because the REY concentration in some coals and coal ashes are equal to or higher than those in conventional deposits (Seredin, 1996; Seredin and Dai, 2012; Seredin *et al.*, 2013; Hower *et al.*, 2015; Anggara *et al.*, 2018, 2019; Rosita *et al.*, 2020).

Indonesia coal consumption is predicted to exceed 200 million tons within the coming decade, and increases due to the government plan on building several more coal -fired power plants (CFFPs) to meet the energy needs. This combustion process produces ashes known as fly ash and bottom ash (FABA). Overall, only 25% of this FABA hasbeen utilized and the rest remains as landfilled solid waste (Sommerville *et al.*, 2013; Blisset*et al.*, 2014). Having the approach of REY recovery as by-product of FABA could alleviate the environmental aspect of CFFP and shifted the paradigm from "dirty" to "clean" coal energy.

According to Seredin and Dai (2012), REY-rich coal could be classified into four genetic types, that are (1) terrigenous, (2) tuffaceous, (3) infiltrational, and (4) hydrothermal processes.Kutai Basin is known as prolific coal deposit in Indonesia, as for the coal resource in Sangatta coal deposit has been reported to be about 6,203 Mt (Friederich and Leuween, 2017). Economically, coal bearing sequences in Kutai Basin are controlled by several tectonic setting and processes, such as NNE-SSW trending axes of Neogenic Mahakam fold belt and Pinang Dome as associated features due to anomalous geothermal gradient that may have been thermally affected (Friederich and Leeuwen, 2017). Recently, a study about maceral composition, physical, and chemical properties of Sangatta Miocene coal has been conducted by Anggara et al. (2014). While Moore and Nas (2013) and Moore et al. (2014) had reported the significance and influence on coal rank and coal bed methane properties in Pinang Dome. However, studies about the effect of Pinang Dome on the concentration of REY in Sangatta coal are still very limited, as such the objective of this study is to examine the REY content of Sangatta coal approaching the Pinang Dome.

Regional Geology

The Middle-Late Miocene Balikpapan Formation was deposited during the overall regressive sequences in Kutai Basin (Moore and Nas, 2013). According to Allen and Chambers (1998), there are three tectonic phases that lead to the development of Kutai Basin. The first, syn-rift sequence from Late Cretaceous-Eocene, is an extensional phase resulting in the opening of some half-graben with NE-SW orientation. The second, post-rift sequence started from Oligocene to Middle Miocene, is a basin infilling and basin subsidence. The third, inversion sequence from Late Miocene-Recent, is the Kuching and Meratus orogeny uplift that occurs across the Kutai Basin resulting elongate structural deformation and followed by erosion.

This tectonic setting and regional geological history used to construct the possibility of the Pinang Dome origin (Friederich and Leeuwen, 2017). Paleogene extensional tectonic phase of Mahakam fold-belt followed by Neogene compression phase and basin inversion, and igneous intrusion may then have followed structural weakness as shown in Figure 1.

Kutai Basin is one of the prolific coal basins in Kalimantan Island, Indonesia, with the total coal resources of 40.67 Gt (Anonym, 2017). Coal-bearing sequences in Kutai Basin are mainly Miocene to Eocene in age in which the coal mined are mostly from Miocene seams. One of the deposits in Kutai Basin is Sangatta deposit with resources of 6,203 Mt. The coal rank in this location increases (nonconcentrically) with lateral rate of about 0.03%/1,000 m (Moore and Nas, 2013), and the huminite reflectance varies from 0.37% to 0.50% (Anggara *et al.*, 2014) towards the SW of Pinang Dome.

SAMPLES AND METHODOLOGY

Six samples were collected from coal cores drilled around Pinang Dome with various depths from 30 to 140 m below the surface within the perimeter of Pinang Dome. The samples were the members of Balikpapan Formation (see Figure 2).



Figure 1. Basins in Kalimantan and location of coal samples used in this study (compiled and modified from Friederich *et al.*, 1999; Moss and Chambers, 1999; Satyana *et al.*, 1999).

Formation	Thickness (m)	Lithological Column	Coal Seam Name	Lithological Characteristics
BALIKPAPAN	E 1500 - 1400 - 1300 - 1200 - 1100 - 1000 - 900 - 800 - 700 -		K1 Kedapat Ma1 Mandili P7 P5 P4 P3 P3 P3 P3 P3 P3 P3 P3 P3 P3 P4 P1 Pinang Mi-1 Middle Sangatta B2 B1 Bintang Prima Prima	Mudstone, sandstone, siltstone, thin coal sequence (additional coal seams continue above this section) Mudstone, siltstone, sandstone, and coal. Main West Pinang coal deposit sequence containing coal seams from the Sangatta to Kedapat seam. Mudstones typically show Ironstone nodules and bands Sandstone beds up to 10 m thick. Dominantly mudstone, siltstone with thick channel sandstone units (10 - 30 m thick). In the South of Pinang Area, the Prima and Bintang seams are significant reserves but these seams thin to the north where the sandstone units become more predominant in the sequence.
?	600 -		Tempudau Pamungkas	
PULAU BALANG	500 - 400 -		North Melawan Bara Mutu Gendeng Panjol Coal seams in this interval have not be named	Coal seam to 0.5 to 2.00 m thick only, usually with high Sulphur content (>10.00%). Fluvial sandstone with coal detritus in upper part of interval.
	300 - 200 - 100 -			Dominantly mudstone and siltstone interval with thin calcareous sandstone beds fine grained thin coralline limestone and bioturbated sandstone in lower part of interval. No Coal of economic significance in this part of the sequence.
PAMALUAN	- 0-		/	Limestone, coralline marker bed at base of coal sequence. Mudstone, fine laminated calcareous sandstone, thin . Limestone bands.

Figure 2. Stratigraphic section of Sangatta coal field. Coals were sampled from Balikpapan Formation (based on Macmillan *et al.*, 2000).

The lithotype, maceral, huminite reflectance, proximate, and ultimate analysis have been conducted for coal characterization by Anggara *et al.* (2014). Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES) was used to determine the oxides of major elements in the coal, including SiO₂, Al₂O₃, CaO, K₂O, Na₂O, Fe₂O₃, and MgO. Inductively coupled plasma mass spectrometry (ICP-MS) was used to determine the trace and rare earth elements in the coal. ICP AES/MS analyses were conducted by ALS Canada Ltd. (Vancouver, Canada) using the fused bead method prior to acid digestion.

Coal petrography was done in accordance with ASTM D2799005a (2005), and maceral classification was conducted following the ICCP System 1994. As for proximate analysis, ASTM Standards D3173-03 (2005), D3174-04 (2005), and D3175-02 (2005) were chosen as the standardized methods. Coal petrography and proximate analysis have been conducted in the Department of Geological Engineering, Universitas Gadjah Mada.

RESULTS

Coal Characteristics

Megascopically, coal samples used in this study are classified into two types, banded bright and nonbanded dull coals. Table 1 shows that maceral composition is dominated by huminite (79.6 - 84.0%), followed by liptinite (6.6 - 8.0%) and inertinite (6.0 - 12.2%). Minerals (0.7 - 2.4%) mostly consist of pyrite and clay. On the other hand, carbonate and quartz are very rarely found. For the rank of coal samples, it is observed that the huminite reflectance (Hr) ranges from 0.37% to 0.50% (as shown in Table 1) indicating that all coals are low rank (lignite to subbituminous).

Based on the results of proximate analysis (Table 1), it shows similarity to Hr and coal rank data. The majority of coal samples have low ash yield of 0.7% - 2.9%db. The ultimate analysis results show that samples in this study are humic coal, with subbituminous to lignite in term of coal

rank. The decrease of H/C and O/C ratios reflects the increase of coal rank confirming the huminite reflectance (%) results.

Coal Geochemistry

Major elements in the coals were determined by ICP-AES and reported as weight percent (wt%) of the oxides. Major elements in Sangatta coal are dominated by SiO_2 , Al_2O_3 , and Fe_2O_3 (Table 2). These results positively relate to petrographic analysis in which silica and aluminum are the major constituents of clay mineral that are the dominant mineral presented in the coal samples.

In this study, REY is classified into three groups, Light (LREY): La Ce, Pr, Nd, Sm; Medium (MREY): Eu, Gd, Tb, Dy, Y; and Heavy (HREY): Ho, Er, Tm, Yb, Lu (Table 2). Based on ICP-MS results, REY concentration is in a wide range from 2.34 ppm to 22.43 ppm. The coal sample with the highest REY concentration was located near the Pinang Dome. The individual concentration of REY in coal samples is shown in Figure 3.

DISCUSSION

Main Type of REY DistributionPatterns

REY abundance increases from La to Eu and decreases from Eu to Lu. Generally, MREY and LREY are found much more abundant than that of HREY (Figure 4). Seredin and Dai (2012) proposed three types of REY enrichment in regard to the comparison of the average REY concentration in Upper Continental Crust (UCC) by Taylor and McLennan (1985). The types are L-Type (La_N/Lu_N> 1), M-Type (La_N/Sm_N< 1, Gd_N/Lu_N> 1), and H-Type (La_N/Lu_N< 1). From these ratios,the studied coal samples have H-Type enrichment and mixed type of L-M enrichment (Figure 5).

Anomaly of Rare Earth Element and Yttrium (REY) in Coal

According to Dai *et al.* (2016), anomaly of enrichment or depletion of redox and non-redox sensitives could be used as geochemical indica-

Table 1. P	etrographic	Analysis,	Proximate,	and Ultimate	Analysis of	Coal Samples
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Sample	RE 1	RE 2	RE 3	RE 4	RE 5	RE 6
Petrographic composition (%)						
Textinite	2.2	1.4	0.2	1.4	0.4	0.4
Ulminite	26	18	23.4	23	18.6	19
Total Telohuminite	28.2	19.4	23.6	24.4	19	19.4
Attrinite	14.4	19	10.6	4.8	14.4	17.4
Densinite	13.8	7.3	9.2	11.8	16	8.8
Total Detrohuminite	28.2	26.3	19.8	16.6	30.4	26.2
Corpohuminite	13.1	27.8	26.4	30	18.2	27.6
Gelinite	12.5	6.1	13.6	9.8	12.2	8.2
Total Gelohuminite	25.6	33.9	40	39.8	30.4	35.8
Total Huminite	82	79.6	83.4	80.8	79.8	81.4
Fusinite	2.4	0.8	0	0.2	0.8	0.4
Semifusinite	2	1.8	1.4	2.8	4	1.8
Funginite	1.8	2.2	2	2.4	2.4	3.2
Secretinite	0	0	0	0	0	0
Macrinite	1.3	0.2	0.2	0	0.8	0.6
Micrinite	0	2.5	0.4	1.4	0	0.6
Inertodetrinite	3.1	3.1	2.6	2.6	4.2	3.2
Total Inertinite	10.5	10.6	6.6	9.4	12.2	9.8
Sporinite	0.5	0	0	0.4	0.4	0
Cutinite	1.6	1.2	2	1.6	2.4	2.8
Resinite	1.3	2	1	1.4	1.2	1.2
Alginite	0	0	0	0	0	0
Liptodetrinite	2	2.4	1.6	1.2	0.2	1
Suberinite	0.9	2.2	3.2	1.2	2.2	2.2
Chlorophyllinite	0	0	0	0	0	0
Exsudatinite	0.4	0.4	0.2	0.4	0.4	0.2
Bituminite	0	0	0	0	0	0
Fluorinite	0	0	0	0	0	0
Total Liptinite	6.7	8	8	6.2	6.8	7.4
Mineral Matter	0.7	1.8	2	3.6	1.2	1.4
<i>Huminite reflectance</i> (%)	0.47	0.47	0.47	0.46	0.5	0.5
Proximate analysis						
Moisture (%)	10.1	12	13.2	20.8	21.8	5
Ash (% db)	0.7	1.1	0.9	2.9	2.3	1.6
Volatile matter (% daf)	47	44.9	45.8	47.1	46.2	40.4
Fixed carbon (% dmmf)	53	55.1	54.3	53.1	53.9	53
Ultimate analysis						
C (%)	77.41	77.26	75.53	80.74	79.67	79.7
H (%)	6	6.09	6.05	7.14	7.09	7.4
N (%)	18	1 77	1.83	1 94	1 78	12

tors of the sediment source, sedimentary environment, tectonic evolution, and post depositional history of coal deposit. The pattern shows that the redox sensitive elements Ce and Eu show a positive anomaly, and nonredox sensitive elements La, Gd, and Y show a negative anomaly (Figure 6). Based on the data, the controlling factor of REY anomalies in coal samples may be affected by mafic source and injection of hydrothermal fluids during peat accumulation. In this study, Eu anomaly is used because of its high value and strong anomaly compared to other elements.

Sample	RE 1	RE 2	RE 3	RE 4	RE 5	RE 6
SiO ₂	0.88	1	1.17	0.91	0.7	0.77
Al_2O_3	0.21	0.16	0.15	0.1	0.41	0.73
Fe ₂ O ₃	0.26	0.44	0.37	0.72	0.18	0.42
CaO	0.39	0.07	0.1	0.1	0.42	0.11
MgO	0.06	0.04	0.06	0.03	0.08	0.03
Na ₂ O	0.65	0.02	0.07	0.03	0.53	0.12
K ₂ O	0.02	0.03	0.02	0.02	0.04	0.03
LOI	97.1	98.1	97.8	98	97.1	97.3
La	0.50	0.50	0.50	1.40	0.60	3.60
Ce	0.90	0.60	0.80	3.40	1.20	8.80
Pr	0.11	0.07	0.08	0.41	0.19	1.13
Nd	0.50	0.30	0.30	1.50	0.60	4.20
Sm	0.09	0.11	0.09	0.24	0.16	0.80
Eu	0.03	0.03	0.03	0.04	0.03	0.17
Gd	0.11	0.05	0.06	0.16	0.17	0.57
Tb	0.02	0.01	0.02	0.01	0.03	0.09
Dy	0.10	0.05	0.09	0.07	0.19	0.49
Y	0.60	0.50	0.50	0.50	1.10	2.10
Но	0.02	0.01	0.02	0.02	0.05	0.08
Er	0.03	0.05	0.03	0.03	0.12	0.18
Tm	0.01	0.01	0.01	0.01	0.02	0.03
Yb	0.08	0.04	0.03	0.03	0.12	0.18
Lu	0.01	0.01	0.01	0.01	0.02	0.01
REY	3.11	2.34	2.57	7.83	4.6	22.43
REO	5.20	4.13	3.05	9.20	5.47	26.41
La/Lu	0.53	0.53	0.53	1.49	0.32	3.84
La/Sm	0.83	0.68	0.83	0.88	0.56	0.68
Gd/Lu	0.93	0.42	0.51	1.35	0.72	4.80
Eu/Eu*	1.39	1.81	1.91	0.95	0.85	1.18
Ce/Ce*	0.87	0.71	0.89	1.02	0.80	0.99
Y/Y*	1.02	1.70	0.90	1.01	0.86	0.80
Туре	Н	Н	Н	L and M	Н	L and M

Table 2. Geochemistry of Coal Samples (units in ppm)

*Notes: H = Heavy, L = Light, M = Medium type of REY



Figure 3. Individual rare earth oxide concentration in coal samples.

Strong positive Eu anomaly could be expected as the influence of hydrothermal fluids at >200°C



Figure 4. REY distribution normalized by Upper Continental Crust (Taylor and McLennan, 1985).

(Bau, 1991; Danielson *et al.*, 1992) during peat accumulation, because the transformation from



Figure 5. Geochemical enrichment type of coal samples.



Figure 6. The distribution pattern of REY geochemical anomalous in Sangatta coal (normalized by Ketris and Yudovich, 2009).

Eu³⁺ to Eu²⁺ not only requires extremely reductive condition but also high temperature as mentioned above (Bau, 1991). Mafic source related to positive Eu anomaly happens in Guxu Coalfield, China, in which the sediment source was mainly derived from tholeiitic basalts of Kangdian Upland (Xiao *et al.*, 2004). It suggests that the REY in coal samples of Sangatta coal field may be derived from mafic source from Pinang Dome.

REY Content

The highest REY concentration is found in RE-6 with the total REY content of 22.43 ppm. This coal sample was taken from the nearest location to the Pinang Dome (Figure 7). Thus, in accordance to the four genetic types of enrichment processes proposed by Seredin and Dai (2012), REY content in Sangatta coal is considered to be of terrigenous type and hydrothermal type. These types are believed to be associated with the formation of igneous diapir of Pinang Dome conducting hydrothermal fluids that might be from Pinang Dome and mafic sediment source



Figure 7. Sampling location overlayed by Bouguer gravity anomaly map of the Pinang Dome and surrounding areas. Data taken by Friederich and Leeuwen (2017).

from the hinterland. The REY enrichment in RE-6 coal is also identified as L-M Type.

The origin and development of Pinang Dome as igneous diapir during peat accumulation will affect the process of peatification and coalification. As the result, it could affect the coal characteristics, coal rank, coal geochemistry, and the abundances of coal REY content. However, the REY content in Sangatta coal has lower concentration than those found in South Sumatra basin (Anggara *et al.*, 2018, 2019).

Conclusions

From this study, it can be concluded that the coal with the highest REY concentration is found in the nearest location to the Pinang Dome. Coal samples in this study are considered to have mixed L-M type of REY enrichment, and show a strong positive Eu anomaly. Those two characteristics indicate the influence of hydrothermal fluids and mafic sediment-source that are associated with the presence of Pinang Dome in the studied area. This work was partly supported by Faculty of Engineering, Universitas Gadjah Mada.

Based on the collected data, the origin of Pinang Dome could much better be understood. The data is in the favour of igneous diapir as the origin of the Pinang Dome which is still hypothetically debatable. Additional data hopefully could shed the light on the origin of Pinang Dome and the geological processes involved in it.

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