Occurrence of Rare-Earth Elements in Selected British Columbian Coal Deposits and their Derivative Products

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Introduction

Coal, an organic sedimentary rock, is reported to contain 86 elements, including 74 trace elements identified using modern analytical techniques (Tang and Huang, 2002). The presence of trace elements in coal is attributed to prevailing conditions in the coal swamp as well as geological activities and atmospheric inputs that would have occurred from time to time during coal deposit formation (Swaine, 1990). Some of the genetic controls that are responsible for the enrichment of trace elements include the source rock, the environment, and the presence of hydrothermal fluid, groundwater and volcanic ash (Dai et al., 2012). Coal with enriched concentrations of valuable trace elements is referred to as metalliferous or mineralized coal (Seredin and Finkleman, 2008). Even though there is no commercial process, metals such as Al, Au, Pt-group metals, V and Zn have been recovered from coal and its byproducts (Bratskaya et al., 2009; Sahoo et al., 2016). Enriched concentrations of rare-earth elements (REE) in coal were first documented in Russian Far East deposits during the early 1990s (Seredin, 1996) and enriched concentrations of REE in coal deposits have been discovered around the world (Birk and White, 1991; Hower et al., 1999; Seredin, 2004; Dai et al., 2007, 2008).

With the emergence of new clean-energy and defense-related technologies, the consumption of REE has increased rapidly (Tse, 2011). For example, it is projected that the demand for Dy will increase by as much as 2600% by 2025 (Chakhmouradian, 2014). In addition, traditional rare-earth ore deposits are depleting quickly and are projected to meet the demand for only the next 15–20 years (Seredin and Dai, 2012). Based on supply and demand, REE are classified as ‘critical elements’ by the United States and the European Union due to their importance in clean energy and defense applications (United States Department of Energy, 2010; European Commission, 2017). The United States National Energy Technology Laboratory (NETL) has identified coal deposits as a potential source of REE and has conducted a prospective analysis of coal deposits using the United States Geological Survey coal database, which contains data on the concentration of REE across the United States coalfields. Furthermore, the United States Department of Energy has provided US$10 million in funding to research projects that demonstrate the techno-economic feasibility of domestic REE separation technologies from coal and/or its byproducts containing a minimum of 300 ppm total REE and concentrating the REE to a level greater than or equal to 2% (by weight) in processed streams (National Energy Technology Limited, 2016).

Even though the literature indicates the presence of REE in some Canadian coal deposits, especially in British Columbia (BC) coalfields, there has been no effort to properly quantify and characterize these deposits and no analysis of extraction techniques (Birk and White, 1991; Goodarzi et al., 2009). The purpose of this ongoing research is to characterize and quantify the REE and their mode of occurrence in BC coal deposits and coal processing products, and to study the possible extraction of these elements. This paper shows the preliminary results from studies conducted on coal samples originating from coal deposits in southeastern BC.

Background

According to the International Union of Pure and Applied Chemistry (IUPAC), REE are a group of 17 elements in the periodic table, including 15 lanthanides and 2 chemically similar transition metals: Sc and Y. Based on supply and demand, these elements are classified into three categories (Seredin and Dai, 2012):

- critical: Nd, Eu, Tb, Dy, Y and Er
- uncritical: La, Sm, Pr and Gd
- excessive: Ce, Ho, Tm, Yb and Lu

Using the atomic number, REE are classified as heavy REE (HREE) and light REE (LREE), with elements from Tb to Lu and Y belonging to the former group and La to Gd and Sc belonging to the latter group (Dai et al., 2008; Moldoveanu et al., 2013; Zhang et al., 2015).

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More than 200 minerals have been identified as containing REE, but the traditional commercial sources of rare-earth elements are (O'Driscoll, 1991)

- bastnaesite, a fluorocarbonate mineral;
- monazite, a LREE phosphate;
- xenotime, a Y-rich phosphate;
- apatite, a Ca fluorophosphate and
- ion-adsorbed clays, an aluminosilicate mineral.

The grade for REE is defined by the content of their oxides (such as lanthanum oxide, La_2O_3) in the deposit. The cutoff grade for a traditional rare-earth resource is approximately 1.5 to 2.0% rare-earth oxide (REO) except for ion-adsorbed clays, for which it is approximately 0.06 to 0.15% REO (Zhang et al., 2015). As noted earlier, the traditional commercial sources are fast depleting and with the increasing demand of REE, the discovery of coal deposits with enriched concentrations of REE have gained special attention in recent years.

Abundance of REE in Coal

Table 1 lists the average concentration of REE in coal from different countries such as the United States, China, the Democratic People's Republic of Korea and Turkey in comparison with average values for the upper continental crust, black shale and world coal on a whole coal basis. The average REE concentration in world coal on a whole coal basis is 68 ppm (Ketris et al., 2009), which is 2.6 times lower than REE in the upper continental crust (179 ppm; Taylor and McClennan, 1985), and the average black shale values are 182 ppm (Ketris et al., 2009). Approximately 80% of the total REE concentration in coal is LREE (Zhang et al., 2015). The average concentration of REE in Chinese coal varies between 101 and 138 ppm (Dai et al., 2008; Zhang et al., 2015), which is 1.5–2 times greater than the world's average. Similar results were observed for Turkish coal (Karayigit et al., 2000; Zhang et al., 2015). The average REE concentration of coal in the United States and the Democratic People's Republic of Korea is close to the world's average. The average REE concentration in world coal on an ash basis is 404 ppm (Seredin and Dai, 2012); however, coal with enriched concentrations of REE on a coal ash basis have been found in coalfields such as in the Sydney Coal Basin, Nova Scotia (72–483 ppm), the coalfields in the Russian Far East (300–1000 ppm) and the Central Appalachian Basin Fire Clay coal bed in the United States (500–4000 ppm; Birk and White, 1991; Seredin, 1996; Hower et al., 1999). The anomalies of REE in coal and distribution patterns are discussed in detail elsewhere (Dai et al., 2016).

Some of the enriched coal (0.68–2.03% REO) have comparable rare-earth concentrations to traditional commercial rare-earth deposits and these coal ashes can be viewed as a potential source for these metals (Seredin and Dai, 2012).

Mode of Occurrence of REE in Coal

Enriched concentrations of REE in coal can be formed under different geological conditions during the entire coal deposit formation process and by various ore-forming processes (Seredin and Finkelman, 2008; Dai et al., 2016). Table 2 shows the four genetic types of REE enrichment in coal: terrigenous, tuffaceous, infiltrational and hydrothermal.

Rare-earth elements can be associated with both organic (Eskenazy, 1987; Seredin, 1996; Wang et al., 2008) and inorganic matter in coal (Eskenazy, 1987; Birk and White, 1991; Seredin, 1996; Hower et al., 1999; Dai et al., 2008), although REE associated with organic matter are considerably less abundant (Eskenazy, 1987; Birk and White, 1991; Seredin, 1996; Zheng et al., 2007). The organic association of the REE in coal is demonstrated by the strong correlation of HREE with organically fixed trace elements like Ge, W and Be and high concentrations of HREE with low-ash coal (Eskenazy, 1982; Eskenazy, 1987; Querol et al., 1995; Seredin, 1996). Trace elements in coal may be associated with organic matter such as carboxylic acid (-COOH), phenolic hydroxyl (-OH), mercapto (-SH) and amino (=NH) groups (Swaine, 1990). Sorption experiments on REE with xylain and humic acid suggested that in the cation exchange process, ions such as Na^+, K^+, Ca^+ and Mg^{2+}, which are bound to COOH and OH^- groups, might have been replaced by REE ions (Eskenazy, 1999).

The mechanism of the organic matter association of REE can also be sorption during coagulation with humic and fulvic acids, as well as the formation of organometallic complexes (Yershov, 1961; Birk and White, 1991; Wang et al., 2008).

On the other hand, the inorganic matter association of REE in coal can be shown by the strong correlation of REE with ash content. In this case, REE minerals are included in accessory minerals, resistate minerals or clay minerals (Eskenazy, 1987; Birk and White, 1991; Zheng et al., 2007). In addition, REE in the Fire Clay coal deposit in the United States have been found to be authigenic in origin (Hower et al., 1999). Finding the percentage of organic and inorganic REE in a coal deposit is essential because it plays a significant role in developing extraction options (Zhang et al., 2015). The present study examines the concentration of REE in coal and their mode of occurrence in select coalfields in BC.

Materials and Methods

An approximately 300 kg run-of-mine sample from five seams were collected at two coal mines in the East Kootenay coalfields, BC. Figure 1 shows the location of East Kootenay coalfields and coal mines. The representative samples were obtained for testing following the stan-

Table 1. Concentration of rare-earth elements (REE; whole coal basis, in ppm) in coal from different countries in comparison with REE contents in coal worldwide, the upper continental crust and black shale. Abbreviations: HREE, heavy rare-earth elements; LREE, light rare-earth elements; ND, no data.

Location of coal	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Y	Ho	Er	Tm	Yb	Lu	Sc	REE	LREE	HREE
Upper continental crust[1]	30	64	7.1	26	4.5	0.88	3.8	0.64	3.5	22	0.8	2.3	0.33	2.2	0.32	11	179	147	32
Black shale[2]	28	58	4.2	33	5.4	1.2	4.7	0.75	3	26	0.52	1.9	0.4	2.8	0.4	12	182	147	36
World[2,7]	11	23	3.5	12	2	0.47	2.7	0.32	2.1	8.4	0.54	0.93	0.31	1	0.2	3.9	72	59	14
United States[3]	12	21	2.4	9.5	1.7	0.4	1.8	0.3	1.9	8.5	0.35	1	0.15	0.95	0.14	ND	62	49	13
China[4]	26	49	5.5	22	4.3	0.9	3.7	0.7	3.1	18	0.7	1.9	0.27	2.1	0.3	ND	138	111	27
China[5]	18	35	3.8	15	3	0.5	3.4	0.52	3.1	9	0.73	2.1	0.34	2	0.32	4	101	83	18
Turkey[5]	21.12	39.24	4.71	16.85	3.18	0.76	3	0.45	2.42	12.76	0.47	1.37	0.21	1.35	0.21	7.92	116	97	19
Democratic People's Republic of Korea[6]	14.5	27.2	2.9	11.1	2.3	0.5	1.4	0.3	2	7.2	0.4	1.1	0.3	1	ND	4.9	77	65	12

[1] From Taylor and McLennan (1985); [2] from Ketris and Yudovich (2009); [3] from Finkelman (1993); [4] from Dai et al. (2008); [5] from Karayigit et al. (2000), Zhang et al. (2015); [6] from Hu et al. (2006); [7] from Seredin and Finkelman (2008)

Table 2. Genetic types of rare-earth element enrichment in coal (Seredin and Finkelman, 2008; Seredin and Dai, 2012).

Genetic type	Mode of rare earth element input	Coalification stage
Terrigenous	Surface waters	Peat-bog stage
Tuffaceous	Leaching of acid and falling alkaline volcanic ash	Peat-bog stage
Infiltrational	Meteoric groundwater	Epigenetic (lignite, sub-bituminous, bituminous-anthracite)
Hydrothermal	Thermal mineral water and deep fluids	Any stage

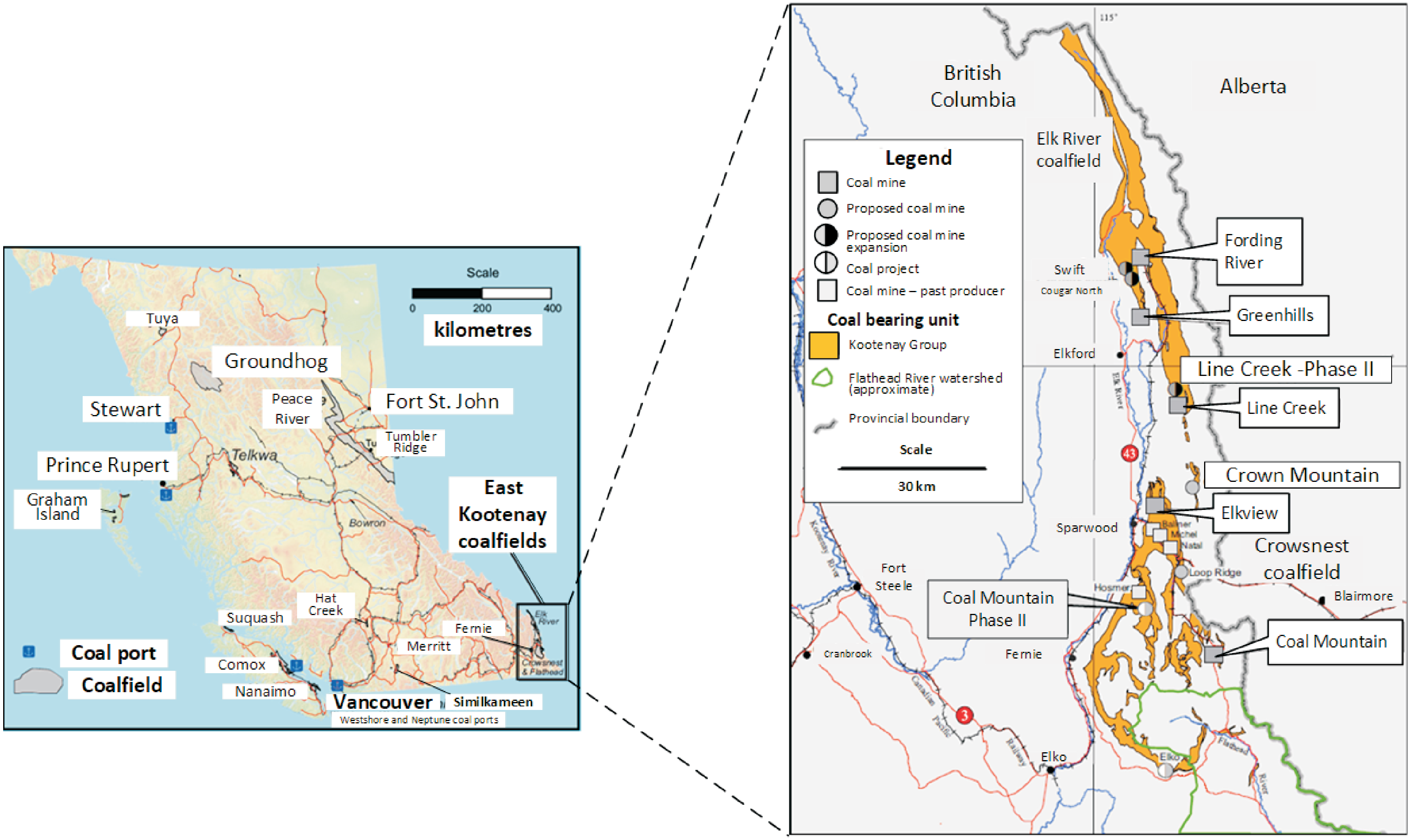

Figure 1. Location of East Kootenay coalfields and coal mines in the southeastern British Columbia (adapted from the BC Ministry of Energy, Mines and Petroleum Resources, 2015, 2016).

dard test procedure ASTM D2013/D2013M-12(2012)[1]. Proximate analysis was conducted on the representative samples in duplicates using the standard methods ASTM D3173/D3173M–17(2017), ASTM D3174–12(2012), ASTM D3175–17(2017) and ASTM D3172–13(2013).

Release Analysis Test

The release analysis test is a technique that is considered to produce an ideal separation curve for the flotation response of any coal. In the release analysis test, as initially outlined by Dell (1964), a flotation test is performed in two stages. In the first stage, coal is floated to achieve maximum recovery, whereas in the second stage, all products from the previous stage are separated into different concentrate products with varying grades and yields. Because products from the release test will have fractions consisting of clean coal (low mineral matter) to tailings (liberated mineral matter) with incremental ash percent, studying REE concentration in these products helps increase understanding of the nature of REE association (organic or inorganic) in the studied samples. For the first stage of release analysis test, a 250 g sample passing 500 µm was mixed with 1800 mL of tap water in a laboratory-scale 2 L Denver flotation cell and conditioned for 5 minutes at 1200 rpm. Then, the coal slurry was subjected to flotation reagents using 50 g/t methyl isobutyl carbinol (MIBC) and 200 g/t kerosene and conditioned for 14 minutes followed by another 50 g/t of frother addition and conditioning for 1 minute. Using an aeration rate of 4–5 L/min of air, concentrate was collected until the froth was barren; if required, extra collector and frother were added to ensure the maximum recovery. In the second stage, concentrate was fractionated into different products using variable impeller speed and aeration rates.

Agglomeration Test

Efficiency of very fine coal recovery from conventional flotation is usually poor (Sastri et al., 1988). The presence of ultrafine slimes or clays also increases the difficulties of processing fine coal, which is ultimately disposed into tailings ponds (Laskowski, 2001). In coal agglomeration, fine coal particles are mixed with the oil under intense agitation, which preferentially covers only fine coal particles and facilitates aggregation of these particles into clusters called agglomerates. Because there is no lower particle size limit for agglomeration, it can be used to recover very fine coals from tailings ponds (Mehrotra et al., 1983).

The presence of REE in agglomerated coal products were also studied in this project. If successful, this agglomeration process may encourage cleaning tailings as a value-added product with REE recovery from the tailings ponds.

In this study, pure diesel was used as a reagent for agglomeration. Reagent concentrations varying from 8 to 20% (by weight) were used for agglomerating a fine coal slurry containing 12% (by weight) coal (sample S3) with a P_{80} (80% of particles finer than the size) of 250 µm. In each test, 500 ml of a 12% coal slurry was conditioned in a high-speed blender at 12 000 rpm for 2 minutes. The reagent (diesel) of required concentration was added and mixing continued for an additional 150 seconds before the mixture was poured onto the sieves to obtain three size fractions: >500 µm (concentrate), <500 µm to >210 µm (middling) and <210 µm (tailings). These products were dried and used for the chemical analysis.

Chemical Analysis of REE and Characterization

For total REE quantification, feed and test product samples (0.2 g) were added to lithium metaborate and lithium tetraborate flux and mixed thoroughly. The samples were then fused in a furnace at 1025°C. Finally, resulting melts were cooled and digested in an acid mixture containing nitric, hydrochloric and hydrofluoric acids. The digested solutions were then analyzed by inductively coupled plasma–mass spectrometry (ICP-MS). The analytical results were corrected for inter-elemental spectral interference. This analysis was conducted by ALS Geochemistry (Vancouver, BC). In this study, REE in coal are expressed as follows: whole coal basis (REE concentration in the coal sample) and ash basis (REE concentration in the ash of the coal sample).

Additional sample characterization was conducted using FEI Quanta 650 ESEM (environmental scanning electron microscope) equipped with scanning energy microscopy with energy dispersive X-ray spectroscopy (SEM-EDX).

Results and Discussion

Results of the as-determined proximate analysis of the samples are shown in Table 3. According to ASTM D388-17(2017), all the feed coal samples (samples S1–S5) are classified as medium volatile bituminous coal. Flotation release analysis tests provided an estimate of ideal separation achievable for the given coal. Samples used for the release tests showed excellent performance with the clean concentrate containing less than 10% ash while recovering 85% of the combustible material. In the case of the agglomeration test, results indicated that selectivity was not possible with a similar amount of ash in the concentrate (S10), middling (S11) and tailings (S12), which can be observed in the Table 3. All five coal samples along with the flotation and agglomeration products and their respective ashes were analyzed for the REE content and the results are shown in Table 4. Table 5 shows the calculated REE values on a whole coal basis.

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[1] *References to the relevant ASTM methods can be found at the end of the 'References' section.*

Table 3. Proximate analysis of the five British Columbia feed coal samples and flotation and agglomeration products (as-determined basis)

Sample ID	Sample name	Moisture (%)	Volatile matter (%)	Ash (%)	Fixed carbon (%)
S1	Feed sample	2.97	21.01	15.93	60.09
S2	Feed sample	1.49	18.5	18.44	61.57
S3	Feed sample	2.19	16.42	39.63	41.76
S4	Feed sample	2.81	23.27	16.93	56.99
S5	Feed sample	1.13	22.26	20.21	56.4
S6	Flotation concentrate	0.99	20.74	6.39	71.88
S7	Flotation tailings	0.9	19.72	18.64	60.74
S8	Flotation concentrate	0.94	20.79	10.82	67.45
S9	Flotation tailings	0.96	19.79	67.01	12.24
S10	Agglomeration concentrate	1.33	23.67	19.45	55.55
S11	Agglomeration middling	1.03	21.83	19.81	57.33
S12	Agglomeration tailings	1.16	21.22	22.56	55.06

The REE content in the samples varied from approximately 240 ppm to 570 ppm (Table 4), and Ce, La, Nd and Y accounted for 80% of the total REE. Among the five feed samples, sample 1 had the highest REE content (~350 ppm) and sample 3 had the lowest content (~270 ppm). Also, among the treated samples, the REE content in flotation concentrate (~569 ppm) is more than the tailings (~340 ppm) and the similar trend was observed for the agglomeration product (~315 ppm) compared to its tailings (~258 ppm). Furthermore, mass balance showed that 60 to 80% of total REE by weight in the coal is present in the tailings and middling streams.

Preliminary Economic Evaluation of BC Coal as Raw Materials for REE

Based on the rare-earth oxide (REO) content, a coal seam with a thickness greater than 5 m may be considered as a potential source of REE if the minimum REO content is above 800–900 ppm in the coal ash formed from combustion (Seredin and Dai, 2012). Assuming a valence state of +2 for Ce and +3 for the remaining REE, REO content for the samples were calculated. Also, using the ash data from Table 3, REE content on a whole coal basis were determined. Total REO and REE (whole coal basis) values are given in Table 6. The REO concentrations in the samples (Table 6) do not meet the cut-off grade as described by Seredin and Dai (2012), but they still represent a significant resource potential. As mentioned earlier, the coal seams in the United States with REE content of more than 300 ppm are still considered as a potential source for developing the extraction process (National Energy Technology Laboratory, 2016). This shows that BC coal deposits are prospective REE sources depending on the advancement in the extraction practices that could be applied for recovery.

One of the important trends observed in this study is that the product with a low ash content has higher REE concentration on an ash basis but lower concentration on a whole coal basis (Table 6), which might be due to organic dilution. It implies that a significant portion of REE are associated with ash components in the samples studied. When organic matter in coal is oxidized during the ashing process, REE associated with the organic matter of the coal reports to the ash, thus increasing the REE content of the ash. These associations can be organic REE compounds or microdispersed REE minerals. Further studies, including sequential extraction and sodium hydroxide extraction of REE from the coal, are necessary to assess the percentage of organic association in the samples.

The outlook coefficient is another factor that can be used to assess the quality of the REE concentration in the coal seam, which can be defined as the ratio between the relative amounts of critical REE in the sample to the relative amounts of excessive REE in the sample (Seredin and Dai, 2012). It can be calculated as

$$C_{outl} = \frac{\dfrac{Sum\ concentrations\ of\ Nd, Eu, Tb, Dy, Er\ and\ Y}{Total\ REE\ concentration}}{\dfrac{Sum\ concentrations\ of\ Ce, Ho, Tm, Yb\ and\ Lu}{Total\ REE\ concentration}}$$

where Coutl is the outlook coefficient. A higher value for this index represents a higher market value of REE in the coal seam because the concentration of critical REE increases with the index, allowing for the REE in coal as a potential source for industrial purposes. The Coutl for world coal is 0.64 (Ketris et al., 2009; Zhang et al., 2015). The calculated values of Coutl for the five coal samples are given in Table 7. The Coutl values for the BC coal samples are near to or more than 1, implying that critical REE concentration is significant and accounts for more than 35% of the total REE. This provides further evidence for BC coal deposits to be potential sources for REE. The correlation between the outlook coefficient and REE content is believed to be an indicator of organic REE; otherwise, the dominant carrier of REE is authigenic minerals (Seredin, 1996;

Table 4. Rare-earth element (REE) content of the five British Columbia feed coal samples and flotation and agglomeration products (ash basis, in ppm).

Sample ID	Sample name	Ce	Dy	Er	Eu	Gd	Ho	La	Lu	Nd	Pr	Sm	Tb	Tm	Y	Yb	REE* (ash basis)
S1	Feed sample	114	9.82	5.93	2.69	10.5	2.04	57.6	0.92	55.2	14.15	13	1.62	0.92	55.6	5.73	349.72
S2	Feed sample	101	8.4	5.13	2.6	8.66	1.79	53.1	0.76	45.4	12.25	9.77	1.73	0.8	58.8	4.81	315
S3	Feed sample	91.3	6.6	4.2	1.7	7.04	1.45	48.5	0.65	41.5	11.15	8.62	1.09	0.6	41.5	3.91	269.81
S4	Feed sample	97.2	7.1	4.66	2.33	7.71	1.55	55	0.66	44.5	11.85	8.92	1.54	0.69	53.6	4.39	301.7
S5	Feed sample	101	9.02	5.47	2.52	8.76	1.9	53.5	0.86	46.8	12.25	10.35	1.74	0.8	59.7	5.2	319.87
S6	Flotation concentrate	184.5	15.95	9.88	4.05	16.05	3.38	97.1	1.65	85.5	22.7	17.95	2.67	1.51	96.3	9.8	568.99
S7	Flotation tailings	110.5	9.67	5.91	2.51	9.48	1.95	57.9	0.94	52.3	13.4	11.15	1.58	0.87	56.7	5.39	340.25
S8	Flotation concentrate	140.5	12.95	7.82	3.24	12.65	2.63	73.7	1.59	66.3	17.4	14.55	2.09	1.18	76.7	7.72	441.02
S9	Flotation tailings	82.5	5.75	3.66	1.61	6.31	1.19	43.6	0.57	37.8	10.1	8.07	0.99	0.53	35.3	3.31	241.29
S10	Agglomeration concentrate	100.5	8.75	5.32	2.51	9	1.83	52.7	0.84	46.3	12.15	10.6	1.62	0.8	56.6	5.19	314.71
S11	Agglomeration middling	95.5	8.15	5	2.25	8.39	1.69	50.9	0.81	44	11.75	9.35	1.53	0.76	53.4	4.91	298.39
S12	Agglomeration tailings	84.2	6.32	3.92	2.07	6.88	1.32	45.7	0.59	38.4	10.15	7.87	1.43	0.57	44.8	3.6	257.82

*REE = Ce + Dy + Er + Eu + Gd + Ho + La + Lu + Nd + Pr + Sm + Tb + Tm + Y + Yb + Sc** + Pm***

**ICP-MS analysis was not done for Sc.

***Pm is extremely rare and generally considered not to exist in nature (Resende and Morais, 2010).

Table 5. Rare-earth element (REE) content of the five British Columbia feed coal samples and flotation and agglomeration products (whole coal basis, in ppm).

Sample ID	Sample name	Ce	Dy	Er	Eu	Gd	Ho	La	Lu	Nd	Pr	Sm	Tb	Tm	Y	Yb	REE* (whole coal basis)
S1	Feed sample	18.16	1.56	0.94	0.43	1.67	0.33	9.18	0.15	8.80	2.25	2.07	0.26	0.15	8.86	0.91	55.72
S2	Feed sample	18.62	1.55	0.95	0.48	1.60	0.33	9.79	0.14	8.37	2.26	1.80	0.32	0.15	10.84	0.89	58.08
S3	Feed sample	36.19	2.62	1.66	0.67	2.79	0.57	19.22	0.26	16.45	4.42	3.42	0.43	0.24	16.45	1.55	106.94
S4	Feed sample	16.46	1.20	0.79	0.39	1.31	0.26	9.31	0.11	7.53	2.01	1.51	0.26	0.12	9.07	0.74	51.08
S5	Feed sample	20.42	1.82	1.11	0.51	1.77	0.38	10.81	0.17	9.46	2.48	2.09	0.35	0.16	12.07	1.05	64.66
S6	Flotation concentrate	11.80	1.02	0.63	0.26	1.03	0.22	6.21	0.11	5.47	1.45	1.15	0.17	0.10	6.16	0.63	36.38
S7	Flotation tailings	20.59	1.80	1.10	0.47	1.77	0.36	10.79	0.18	9.75	2.50	2.08	0.29	0.16	10.57	1.00	63.41
S8	Flotation concentrate	15.20	1.40	0.85	0.35	1.37	0.28	7.97	0.17	7.17	1.88	1.57	0.23	0.13	8.30	0.83	47.70
S9	Flotation tailings	55.28	3.85	2.45	1.08	4.23	0.80	29.22	0.38	25.33	6.77	5.41	0.66	0.36	23.66	2.22	161.69
S10	Agglomeration concentrate	19.55	1.70	1.03	0.49	1.75	0.36	10.25	0.16	9.01	2.36	2.06	0.32	0.16	11.01	1.01	61.22
S11	Agglomeration middling	18.92	1.61	0.99	0.45	1.66	0.33	10.08	0.16	8.72	2.33	1.85	0.30	0.15	10.58	0.97	59.12
S12	Agglomeration tailings	19.00	1.43	0.88	0.47	1.55	0.30	10.31	0.13	8.66	2.29	1.78	0.32	0.13	10.11	0.81	58.17

*REE = Ce + Dy + Er + Eu + Gd + Ho + La + Lu + Nd + Pr + Sm + Tb + Tm + Y + Yb + Sc** + Pm***

Table 6. Rare-earth oxide (REO) content and rare-earth element (REE; whole coal and ash basis) content for British Columbia coal samples and flotation and agglomeration products (in ppm).

Sample ID	Sample name	REE (ash basis)	REE (whole coal basis)	Total REO
S1	Feed sample	349.72	55.72	420.88
S2	Feed sample	315	58.08	379.93
S3	Feed sample	269.81	106.94	324.88
S4	Feed sample	301.7	51.08	363.72
S5	Feed sample	319.87	64.66	385.67
S6	Flotation concentrate	568.99	36.38	685.34
S7	Flotation tailings	340.25	63.41	409.75
S8	Flotation concentrate	441.02	47.7	531.2
S9	Flotation tailings	241.29	161.69	290.42
S10	Agglomeration concentrate	314.71	61.22	379.3
S11	Agglomeration middling	298.39	59.12	359.64
S12	Agglomeration tailings	257.82	58.17	310.77

Table 7. Outlook coefficient (C_{outl}) for British Columbia coal samples and flotation and agglomeration products. Abbreviation: REE, rare-earth elements.

Sample ID	Sample name	C_{outl}	Critical REE (ppm)
S1	Feed sample	1.06	130.86
S2	Feed sample	1.12	122.06
S3	Feed sample	0.99	96.59
S4	Feed sample	1.09	113.73
S5	Feed sample	1.14	125.25
S6	Flotation concentrate	1.07	214.35
S7	Flotation tailings	1.08	128.67
S8	Flotation concentrate	1.1	169.1
S9	Flotation tailings	0.97	85.11
S10	Agglomeration concentrate	1.11	121.1
S11	Agglomeration middling	1.1	114.33
S12	Agglomeration tailings	1.07	96.94

Hower et al., 1999) and there is no correlation observed for the BC coal samples that implies the REE may originate from authigenic minerals.

Correlation Analysis

Table 8 shows the correlation coefficients of REE (ash basis) with other elements (ash basis) and ash in the coal samples. Table 9 shows the correlation coefficients for REE (whole coal basis) with other elements (whole coal basis). A strong correlation between ash and REE content calculated on a whole coal basis is observed (r = +0.95 to +0.99), indicating the presence of REE mineral phases in the coal samples. One such rare-earth mineral in these types of metalliferous coal is zircon, which can originate from volcanic ash or authigenic minerals and can be identified by the enrichment of Hf, Th, U, Y and HREE (Finkelman, 1981; Seredin, 2004). A compelling correlation between Hf, Th, U, Y and other REE implies that zircon is one of the source minerals of REE and an indication of volcanic ash input of REE into these coal samples. Elemental analysis results using ICP-MS proved the presence of zircon in the samples. Also, volcanic ash is believed to be source for the tonsteins in the Mist Mountain Formation, which further validates the inference made in this study (Grieve, 1993).

Weak negative correlation between ash and REE content on an ash basis (r = –0.58 to –0.78), however, highlights that a small portion of REE in the samples are associated with coal organic matter due to the organic affinity of certain REE with humic acid. A similar trend has been identified in the Sydney Coal Basin, Nova Scotia (Birk and White, 1991), which further supports the inferences made in this study. The presence of a strong correlation between REE and W, which is believed to be organically fixed in coal, is additional evidence for the presence of organically associated REE in the coal.

Figure 2 shows a similar pattern between REE content normalized to upper continental crust values for all the feed and product samples. This may indicate the REE in the ash material in the organic matrix, partings, roof and floor ma-

Table 8. Correlation coefficients of rare-earth elements (REE; ash basis) with other elements in British Columbia coal samples.

Correlation	Ash	REE (ash basis)	Ga	Hf	Nb	Ta	Th	U	V	W	Zr	Ce	Dy	Er	Eu	Gd	Ho	La	Lu	Nd	Pr	Sm	Tb	Tm	Y	Yb
Ash	1																									
REE (ash basis)	–0.63	1																								
Ga	–0.62	0.99	1																							
Hf	–0.63	0.99	0.99	1																						
Nb	–0.53	0.92	0.93	0.93	1																					
Ta	–0.31	0.85	0.84	0.8	0.75	1																				
Th	–0.63	0.95	0.95	0.97	0.93	0.68	1																			
U	–0.58	0.94	0.95	0.97	0.96	0.69	0.98	1																		
V	–0.4	0.71	0.74	0.74	0.93	0.52	0.79	0.85	1																	
W	–0.58	0.89	0.91	0.86	0.86	0.8	0.82	0.86	0.69	1																
Zr	–0.62	0.99	0.99	1	0.94	0.8	0.97	0.97	0.76	0.87	1															
Ce	–0.59	1	0.99	0.99	0.92	0.87	0.94	0.94	0.72	0.9	0.99	1														
Dy	–0.66	0.99	0.97	0.97	0.85	0.83	0.94	0.91	0.61	0.87	0.97	0.98	1													
Er	–0.65	0.99	0.98	0.98	0.88	0.84	0.95	0.93	0.66	0.89	0.98	0.99	1	1												
Eu	–0.76	0.97	0.96	0.96	0.86	0.75	0.93	0.9	0.65	0.86	0.95	0.95	0.97	0.97	1											
Gd	–0.65	0.99	0.98	0.98	0.86	0.85	0.93	0.9	0.62	0.87	0.97	0.99	0.99	1	0.97	1										
Ho	0.66	0.99	0.98	0.98	0.87	0.82	0.95	0.92	0.64	0.87	0.98	0.99	1	1	0.97	1	1									
La	0.6	0.99	1	0.99	0.95	0.85	0.95	0.96	0.77	0.9	0.99	1	0.97	0.98	0.95	0.97	0.98	1								
Lu	–0.59	0.95	0.93	0.94	0.82	0.8	0.91	0.89	0.6	0.83	0.94	0.95	0.97	0.97	0.92	0.96	0.96	0.94	1							
Nd	–0.59	1	0.99	0.98	0.9	0.89	0.93	0.92	0.68	0.9	0.98	1	0.98	0.99	0.95	0.99	0.99	0.99	0.95	1						
Pr	–0.58	1	0.99	0.98	0.92	0.88	0.94	0.93	0.71	0.89	0.99	1	0.98	0.99	0.95	0.99	0.99	0.99	0.95	1	1					
Sm	–0.6	0.98	0.96	0.95	0.83	0.89	0.89	0.86	0.57	0.86	0.94	0.98	0.98	0.98	0.95	0.99	0.98	0.95	0.95	0.99	0.98	1				
Tb	–0.78	0.94	0.94	0.95	0.86	0.67	0.94	0.91	0.67	0.85	0.95	0.92	0.94	0.94	0.99	0.94	0.95	0.93	0.89	0.91	0.92	0.9	1			
Tm	–0.66	0.99	0.98	0.98	0.87	0.83	0.94	0.92	0.64	0.87	0.98	0.99	1	1	0.98	1	1	0.98	0.96	0.99	0.99	0.98	0.95	1		
Y	–0.74	0.97	0.97	0.98	0.9	0.71	0.97	0.95	0.71	0.86	0.98	0.96	0.97	0.97	0.99	0.96	0.98	0.96	0.93	0.95	0.95	0.93	0.99	0.98	1	
Yb	–0.65	0.99	0.98	0.98	0.88	0.82	0.95	0.93	0.65	0.87	0.98	0.99	1	1	0.97	1	1	0.98	0.97	0.99	0.99	0.98	0.95	1	0.98	1

Table 9. Correlation coefficients of rare-earth elements (REE; whole coal basis) with other elements in the British Columbia coal samples.

Correlation	Ash	REE (whole coal basis)	Ga	Hf	Nb	Ta	Th	U	V	W	Zr	Ce	Dy	Er	Eu	Gd	Ho	La	Lu	Nd	Pr	Sm	Tb	Tm	Y	Yb
Ash	1																									
REE (whole coal basis)	0.993	1																								
Ga	0.997	1	1																							
Hf	0.952	0.98	0.97	1																						
Nb	0.969	0.97	0.97	0.95	1																					
Ta	0.799	0.84	0.83	0.81	0.82	1																				
Th	0.992	0.99	0.99	0.98	0.98	0.78	1																			
U	0.988	0.99	0.99	0.98	0.98	0.78	1	1																		
V	0.93	0.92	0.93	0.9	0.98	0.73	0.94	0.95	1																	
W	0.97	0.97	0.97	0.91	0.94	0.83	0.96	0.96	0.9	1																
Zr	0.904	0.94	0.93	0.99	0.93	0.79	0.94	0.95	0.89	0.86	1															
Ce	0.992	1	1	0.98	0.98	0.85	0.99	0.99	0.93	0.97	0.94	1														
Dy	0.973	0.99	0.98	0.98	0.95	0.84	0.98	0.98	0.88	0.95	0.95	0.99	1													
Er	0.98	1	0.99	0.98	0.96	0.84	0.99	0.98	0.9	0.96	0.95	0.99	1	1												
Eu	0.99	0.99	0.99	0.95	0.95	0.8	0.99	0.98	0.89	0.96	0.9	0.98	0.98	0.98	1											
Gd	0.986	1	0.99	0.98	0.96	0.85	0.99	0.98	0.9	0.96	0.94	1	1	1	0.99	1										
Ho	0.976	0.99	0.99	0.99	0.95	0.82	0.99	0.98	0.89	0.95	0.96	0.99	1	1	0.98	1	1									
La	0.994	1	1	0.97	0.98	0.84	0.99	0.99	0.94	0.97	0.94	1	0.98	0.99	0.98	0.99	0.99	1								
Lu	0.947	0.97	0.97	0.97	0.93	0.86	0.96	0.96	0.86	0.93	0.95	0.97	0.99	0.99	0.95	0.98	0.98	0.97	1							
Nd	0.99	1	1	0.97	0.97	0.86	0.99	0.99	0.92	0.97	0.94	1	0.99	1	0.98	1	0.99	1	0.98	1						
Pr	0.991	1	1	0.98	0.97	0.85	0.99	0.99	0.93	0.97	0.94	1	0.99	0.99	0.98	1	0.99	1	0.97	1	1					
Sm	0.982	1	0.99	0.97	0.95	0.87	0.98	0.97	0.89	0.96	0.93	0.99	0.99	1	0.98	1	0.99	0.99	0.98	1	1	1				
Tb	0.985	0.98	0.98	0.94	0.94	0.74	0.98	0.97	0.89	0.95	0.89	0.97	0.97	0.97	0.99	0.97	0.97	0.97	0.93	0.97	0.97	0.97	1			
Tm	0.977	0.99	0.99	0.98	0.95	0.85	0.98	0.98	0.89	0.95	0.95	0.99	1	1	0.98	1	1	0.99	0.98	0.99	0.99	1	0.97	1		
Y	0.987	0.99	0.99	0.97	0.96	0.76	1	0.99	0.91	0.95	0.94	0.99	0.99	0.99	0.99	0.99	0.99	0.99	0.96	0.98	0.99	0.98	0.99	0.99	1	
Yb	0.974	0.99	0.99	0.98	0.95	0.84	0.99	0.98	0.89	0.95	0.96	0.99	1	1	0.98	1	1	0.99	0.99	0.99	0.99	0.99	0.96	1	0.99	1

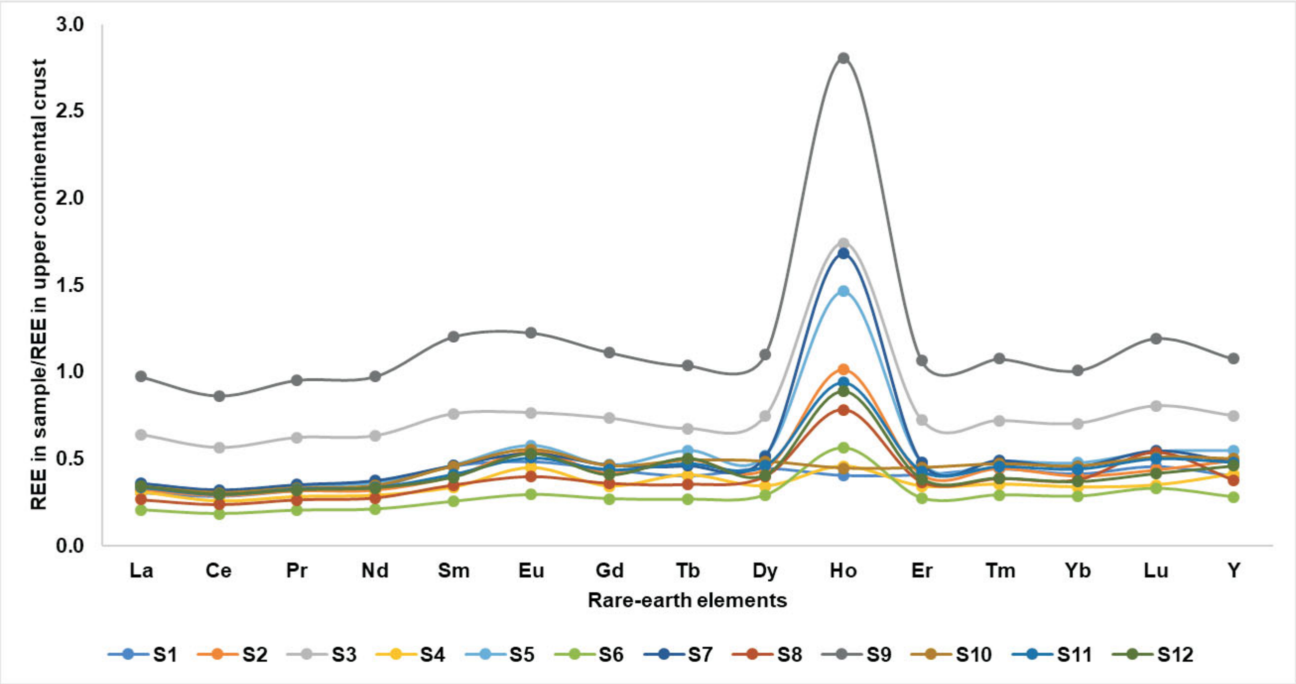

Figure 2. Upper continental crust–normalized rare-earth element (REE) content in British Columbia coal samples.

terial have the same source. The REE sites in the samples were identified using SEM-EDX.

Figure 3 shows grains that contain REE in the sample S3. Based on the elements found using SEM-EDX, it can be inferred that some of the REE are associated with aluminosilicate minerals, which may be clay, although further analysis is required. In addition, REE strongly correlate with U (r > +0.86 on ash basis; r > +0.96 on whole coal basis) and Th (r > +0.89 on ash basis; r > +0.98 on whole coal basis). This suggests that one of the REE mineral phases could be monazite.

Conclusions

Five coal samples from BC coal deposits were tested for the presence of REE and it was found that total REE concentration on an ash basis varied from 240 to 570 ppm. It was inferred from the data that REE in the coal samples are associated with both the organic and inorganic portions of the coal constituents. The highest concentration was reported in the flotation product with low ash content, indicating that some of the REE are associated with the organic matter of the coal. Additionally, correlation analysis showed a significant amount of organic matter associated with REE is due to the organic affinity of REE with humic acid. Total REE concentration versus the outlook coefficient did not, however, show any pattern suggesting the presence of authigenic REE minerals either. Rare-earth element content normalized to the upper continental crust indicated a similar source for all the samples and one of the possible REE sources identified as volcanic ash. The SEM-EDX and chemical analysis data suggested that minerals that contain REE in the coal samples could be monazite and zircon and some of the REE could also be associated with aluminosilicate minerals. Further studies will include sequential extraction, X-ray diffraction analysis and detailed SEM-EDX, required to estimate the quantity of REE associated

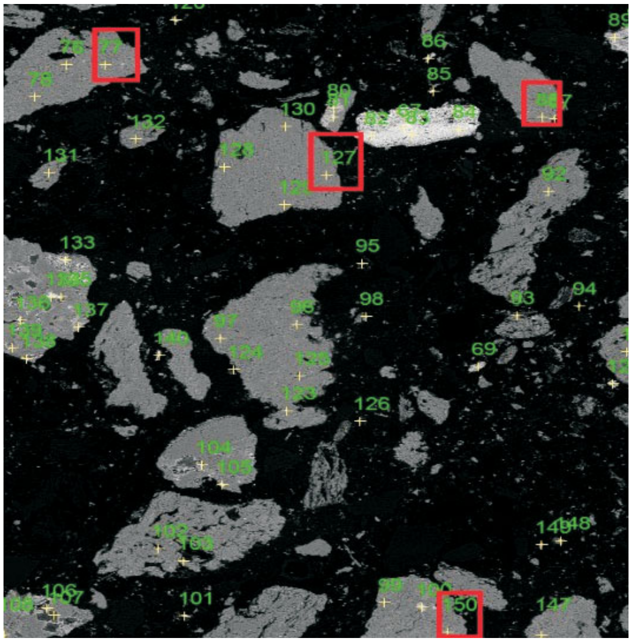

Figure 3. Rare-earth element (REE) sites in sample S3 (200 times magnification); the elements observed in the highlighted grains are as follows: spot 77: Dy, Gd, K, O, P, Si, Y; spot 88**:** Dy, Fe, K, O, P, Si, Ti; spot 127: Al, Ba, Ca, Ce, Fe, K, O, P, Si, Ti; spot 150: Al, Ce, K, O, P, Si.

with organic matter and inorganic matter of the coal samples.

With the development of extraction techniques for REE in coal, these elements will be extracted as byproducts of coal mining, which strengthens the brownfield operations by increasing profitability and possible green credits as these REE are used in the clean energy technologies. In case of the greenfield operations, the possibility of extracting REE from coal will increase the competitiveness of the deposits and its feasibility for actual mining and extraction.

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