

# Coal Fly Ash as Contaminant Barrier for Reactive Mine Tailings

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## 1. INTRODUCTION

The acid mine drainage (AMD) generated from reactive mine tailings has been a major environmental challenge for the mining industry. The oxidation of reactive sulphide minerals and consequent release of acidic water in tailings impoundments can last for decades or even centuries. AMD typically has elevated concentrations of heavy metals and other contaminants, which can severely affect the surface water and groundwater systems. Once these chemical reactions initiate, they are very difficult to stop under the natural conditions. Furthermore, the initiation of the reactive mine tailings depends on many factors. It was reported that many rock piles started to produce acidic seepages many years after the piles were established.

A survey conducted by the Government of Canada in 1991 indicated that the Canadian mineral industry generates 1,000,000 tonnes of waste rock and 950,000 tonnes of tailings per day, totalling 650 million tonnes of waste mine rock per year.<sup>1</sup> Most of these wastes have the potential to produce AMD. The collective liability from the sulphide containing mine wastes was anticipated to be between \$2 and \$5 billion in Canada.<sup>2</sup>

The traditional contaminate barrier techniques commonly used in mine tailings disposal sites include covers and liners made of low permeability materials, such as compacted clay soil, geomembrane and their combinations. Water cover is considered a preferred choice when a water source is available. The objectives of most cover designs are to inhibit the influx of atmospheric oxygen into tailings, thus minimizing oxidation of reactive tailings by reducing infiltration.

A contaminant barrier system in a mine tailings disposal site can be enhanced when they are constructed with materials that have potentials of reducing the seepage velocity of AMD generated from reactive tailings. Furthermore, it is also desirable that the materials can result in immobilization of heavy metals and other toxic ions in discharge. Coal fly ash from coal-fired power generation plants is a potential candidate for such a material. Since coal fly ash generates alkaline leachate upon in contact with water, with proper engineering design, it can be utilized in mine tailings management.

There have been many related research activities in this regard. The research conducted by Nhan et al. (1996)<sup>3</sup> found, with the mixture of lime dust and bentonite, coal fly ash

liner has the hydraulic conductivity of  $4.3 \pm 1.6 \times 10^{-8}$  m/s. They also found the composite liner reduced the dissolved metals in the leachate. The result of a case study by Mollamahmutoglu and Yilmaz (2001)<sup>4</sup> is consistent with the conclusions of Nhan et al., who further reported that in terms of hydraulic conductivity, the optimum mass ratio of bentonite-ash is 1:5. Palmer et al. (2000)<sup>5</sup> introduced a mixture of Class F and Class C ashes and coarse aggregate can be compacted to hydraulic conductivities near or less than  $10^{-7}$  cm/s under the optimal water content. Palmer et al. also pointed out that the freeze-thaw and wet-dry cycles have limited impact on the changes in the hydraulic conductivity in the field test. Their laboratory and field test showed heavy metal concentrations in pore water did not exceed the Wisconsin groundwater quality standards.

Pranshanth et al. (2001)<sup>6</sup> indicated that the pozzolanic coal fly ashes encourage the formation of gelatinous compounds which block the void space among solid particles. This is the main cause of the hydraulic conductivities decrease of compacted coal fly ash. With more reactive silica and free lime content presented in coal fly ash, the pozzolanic reactivity can be enhanced.

Various applications of coal fly ash in hydraulic barriers were studied by researchers. Thiede and Werner (1996)<sup>7</sup> obtained encouraging result in the field experiments of using coal fly ash and Cu mining residues to replace bentonite and HDPE foils as the base liner and cover, respectively, for a disposal site. Cokca and Yilmaz (2004)<sup>8</sup> used the combination of rubber, bentonite and coal fly ash. The composite material had the hydraulic conductivity of less than  $10^{-9}$  m/s and the leachate was found non-hazardous. Beeghly (1996)<sup>9</sup> introduced the method of mixing the coal fly ash, lime, calcium sulphite and calcium sulphate sludge to initiate a cementitious chemical reaction in the construction of an impermeable pond liner.

One of the benefits of using coal fly ash in contaminate barrier applications is capable of consuming large volumes of fly ash without adversely affecting the environmental or potential land occupation. This approach can be economically attractive especially in the regions near the coal-fired power plants.

In this paper, the engineering properties of two coal fly ashes are investigated, followed by column leaching tests with water and AMD permeation. The results are presented in terms of the critical hydraulic gradient to generate seepage flow, the hydraulic conductivity, pH, and contaminant concentrations in the seepage flow. The data can provide insight for utilization of coal fly ash as contaminate barriers for AMD generating tailings.

## 2. ENGINEERING PROPERTIES OF MATERIALS

### 2.1 Characterization of coal fly ashes

Two coal fly ashes, Atikokan coal fly ash (AFA hereafter) and Nanticoke coal fly ash (NFA hereafter), were studied in the research. The fly ashes are by-products of Atikokan and Nanticoke coal-fired power plants owned and operated by Ontario Power Generation (Fig.1). Table 1

summaries the physical properties, chemical compositions, detected minerals, and carbonates of both fly ashes. Due to the similar thermal history and electrostatic precipitation collection method, Atikokan fly ash and Nanticoke fly ash have similar grain size distributions. They are fine powdery materials with the comparable particle size distribution of a silty soil, see Table 1.I. The specific gravity of AFA is 2.69; while that of NFA is 2.37. The characteristic difference of the fly ashes is due to the combination of various factors such as the source of coal, gradation, particle shape, and chemical composition. The lighter specific gravity may be related to the high carbon residual in the NFA, see Table 1.II. From Harvard compaction tests, the optimum water content of AFA (11%) is much less than NFA (25%), as shown in Table 1.I. The corresponding maximum dry densities are 1.925 g/cm<sup>3</sup> (AFA) and 1.275 g/cm<sup>3</sup> (NFA), respectively. Table 1.II presents the major oxides analysis results from X-ray fluorescence (XRF) of AFA and NFA. The calcium oxide (CaO), which is the major source of alkalinity of coal fly ash, counts for contents 15.66% and 11.53% for AFA and NFA, respectively. According to the specification of Canadian Standards Association (CSA A-3000-98)<sup>10</sup>, both AFA and NFA are Class CI fly ashes and have considerable neutralization capacity to acidic mine drainage. Another notable value in Table 1.II is loss on ignition (LOI), which presents the residual carbon in the coal fly ash. The LOI of NFA (8.05%) is much higher than AFA (0.84%), which may implies that the latter has experienced a more complete combustion process. Table 1.III presents the minerals identified in AFA and NFA by X-ray diffraction (XRD) analysis. Both AFA and NFA show poorly crystalline structure. Only traces of quartz (SiO<sub>2</sub>) are identified in both samples. Mullite (Al<sub>6</sub>Si<sub>2</sub>O<sub>13</sub>) may present in NFA. Amorphous materials are dominant in both coal fly ash samples tested. As shown in Table 1.IV, a small amount of carbonates was identified in both AFA and NFA, which is the secondary source of alkalinity of coal fly ash.

## 2.2 Properties of Acid Mine Drainage (AMD)

AMD is generated during the oxidation of reactive sulphide mine tailings. The AMD used in the experimental study was collected from the Sudbury mine tailings disposal site in Northern Ontario, Canada. Table 2 summarized the results of chemical analysis of AMD. The AMD has pH 3.77, and electrical conductivity 1.98 mS/cm. The results of titration tests indicate high concentrations of Cl<sup>-</sup> and SO<sub>4</sub><sup>2-</sup>, which are the only anions detected from the AMD. The results of inductively coupled plasma (ICP) analysis indicated that Ca<sup>2+</sup>, Na<sup>+</sup>, Mg<sup>2+</sup>, K<sup>+</sup>, Ni<sup>+</sup>, and Al<sup>3+</sup> are the dominant dissolved metals in the AMD. None of the concentrations of hazardous elements in the AMD exceeds the Leachate Quality Criteria of MOE (2000)<sup>11</sup>.

## 3. EXPERIMENTAL METHODS AND APPARATUS

To study the long-term and short-term leaching behaviours of AFA and NFA, column leaching tests were carried in laboratory. The purposes of column leaching tests are:

- To measure the break through hydraulic gradient, i.e. the hydraulic gradient that is required to generate seepage flow in the compacted coal fly ash samples;
- To measure the seepage flow and in turn to calculate the hydraulic conductivity of compacted coal fly ash samples with known density;
- To collect leachate permeated through the coal fly ash samples and monitor the leachate chemical changes versus time.

Based on the results, a preliminary assessment of the feasibility of using coal fly ash as an AMD barrier can be made.

Two sets of experimental apparatus were used in the column leaching tests: fixed-wall permeater and constant head permeater. In the former device, see schematic diagram in Fig. 2(a), a seepage flow was forced to pass the coal fly ash compacted in the column at a constant flow rate (Fernandez and Quigley (1985)<sup>12</sup>). In the latter device, i.e., the constant head permeater (see Fig. 2(b)), the coal fly ash compacted in the column is permeated under a constant total head. To prevent erosion of AMD to the permeaters, all wet parts of both permeaters are made of chemically inert materials, i.e. stainless steel and polymerized vinyl. Details of the experimental apparatus can be found in Wang (2004)<sup>13</sup>.

Two series of column leaching tests were conducted to assess the long-term and short-term behaviours of the coal fly ash samples permeated with water and AMD. In the first test series, AFA samples were compacted into two fixed-wall permeaters at the optimum moisture content (Table 1.I), and permeated with water and AMD, respectively. In the second test series, NFA samples were compacted into two constant head permeaters and permeated by water and AMD, respectively. To make the results from the two test series comparable, the results are presented in terms of the progressive pore volume, which is defined as the total quantity of flow normalized by the initial pore volume of the coal fly ash sample compacted in the testing cells.

The 1<sup>st</sup> test series was carried out under a flow rate much faster than what is anticipated in-situ. The results, therefore, can be used to assess the effect of long term leaching of AMD on the coal fly ash. In the 1<sup>st</sup> test series, 60 pore volumes AMD were permeated through the compacted coal fly ash samples over a period of 92 days. In the 2<sup>nd</sup> test series, the flow rate was much lower (12 pore volumes in 217 days) so that the chemical reactions and consequent precipitation and cementation were allowed to complete.

#### 4. RESULTS AND DISCUSSION

The results of discussion of the permeation tests are discussed in terms of four aspects, i.e., the break-through hydraulic gradient, the hydraulic conductivity, the pore fluid pH and ionic concentrations. All of these properties are presented as a function of progressive pore volumes, which is in turn a function of time.

##### 4.1 Break through hydraulic gradient

Figure 3 presents the changes of hydraulic gradients during the first 5 pore volumes of permeation by water and AMD through compacted AFA samples. It can be observed from the figure that the initial hydraulic gradient for water to break-through the compacted AFA sample was about 17. However, the initial hydraulic gradient for AMD to break-through was 270 – 280 under the otherwise identical conditions, more that 10 times higher than that for water.

The similar results were observed in the 2<sup>nd</sup> test series, as shown in Fig. 4. The initial hydraulic gradient for water to break-through the compacted NFA sample was 17, very similar to the results of the 1<sup>st</sup> test series (Fig. 3). The initial break through hydraulic gradient for AMD was

23. However, the flow stopped within 1 pore volume of seepage. The hydraulic gradient to maintain a steady seepage flow on the NFA sample was 153. Again, the break-through hydraulic gradient of AMD for the compacted NFA sample is near one order of magnitude higher than that of water.

The results showed that both coal fly ash, AFA and NFA, had considerable resistance to seepage of AMD. Upon in contact with AMD, chemical reactions are triggered between AMD - an acidic solution - and coal fly ash solids. A review of the chemical reactions has been carried out in Wang (2004)<sup>13</sup>. The coal fly ash behaved as an AMD barrier mainly due to the free lime (CaO). The precipitation from chemical reactions clogs the flow paths in the compacted coal fly ash, which leads to the reduced seepage flow velocity. Therefore, it is anticipated that with proper engineering design, coal fly ash can act as a hydraulic barrier of AMD on reactive mine tailings disposal sites.

#### 4.2 Hydraulic conductivity

Figure 5 presents the hydraulic conductivities of compacted AFA samples permeated with water and AMD, respectively, in the 1<sup>st</sup> test series. The figures shows that in the first 20 pore volumes of permeation, the hydraulic conductivity of both water and AMD permeated samples fluctuated significantly. The effects of permeants to the hydraulic conductivity of coal fly ash samples can be observed after 40 pore volumes of permeation. For the water permeated AFA sample, the hydraulic conductivity showed a slight increase and approached to  $10^{-5}$  cm/s from the initial  $10^{-6}$  cm/s. On the other hand, when the AFA sample was permeated with AMD, the hydraulic conductivity decreased about two orders in magnitude to  $2.5 \times 10^{-8}$  cm/s from the initial  $10^{-6}$  cm/s within 50 pore volumes.

Figure 6 presents the hydraulic conductivity of water and AMD permeated NFA samples in the 2<sup>nd</sup> test series. The results are consistent with those in the 1<sup>st</sup> test series. The hydraulic conductivity of NFA permeated with water remained fairly constant with time. The magnitude of hydraulic conductivity, i.e.  $10^{-6}$  cm/s, is consistent with data from OPG (Chan 1990)<sup>14</sup>. When the compacted NFA sample was permeated with AMD, the hydraulic conductivity progressively decreased with time, reached approximately  $10^{-9}$  cm/s after about 12 pore volume of permeation, compared to the initial value of  $10^{-6}$  cm/s. The hydraulic conductivity of NFA decreased more than 3 orders of magnitude when the sample was permeated by AMD.

The decrease of hydraulic conductivity is another effect of chemical reactions between AMD and coal fly ash solids. The resulted precipitation clogs the flow path in compacted coal fly ash samples, leading to the drop in the hydraulic conductivity. Furthermore, the cementation formed due to these reactions enhances the stability and strength the compacted fly ash. The typical hydraulic conductivity of silts is in the range of  $10^{-7} \sim 2 \times 10^{-3}$  cm/s (Domenico and Schwartz, 1998)<sup>15</sup>. This is consistent with the results of both AFA and NFA permeated with water, in which the hydraulic conductivities were measured within the range of  $10^{-5} \sim 10^{-6}$  cm/s. However, it was observed in two coal fly ash samples tested that when permeated with AMD, the hydraulic conductivities reduced with time. The decreases were about 2 ~ 3 orders of magnitude as compared with those permeated with water. As a result, the hydraulic conductivity of compacted

coal fly ash can be as low as  $10^{-9}$  cm/s, which is comparable to that of compacted clay. As evidenced, the coal fly ash may be beneficial in applications such as contaminate barriers of reactive mine tailings.

It should be noted that the two order decrease in the hydraulic conductivity of the compacted AFA sample (the 1<sup>st</sup> test series) was achieved after the permeation of 50 pore volume AMD, as shown in Fig. 5. The same order of magnitude decrease in the hydraulic conductivity was recorded within only 10 pore volumes AMD permeation in the NFA sample in the 2<sup>nd</sup> test series, see Fig. 6. This is likely related to the velocity of the seepage flow. The precipitation as a result of chemical reactions is accumulated more quickly under a slower seepage flow, such as the case of the 2<sup>nd</sup> test series. On the other hand, a more rapid seepage flow, as in the case of the 1<sup>st</sup> test series, will flush part of the precipitation out from the compacted fly ash sample.

#### 4.3 pH of effluent – acid neutralization capacity

The pH of effluents from all permeaters was monitored during experiments, as shown in Figs. 7 and 8. From Fig. 7, it is noted that the effluent pH from the compacted AFA samples permeated with water and AMD showed no significant difference within the first 40 pore volumes. After that, the effluent pH of AFA permeated with water and AMD both decreased, and the latter was noticeably lower at the end of permeation (51 pore volumes). The final pH of both effluents, however, remained alkaline.

The effluent pH in the 2<sup>nd</sup> test series shows a similar trend, see Fig.8. The effluent pH values from compacted NFA samples permeated with water and AMD were consistent during the first 5 pore volumes. Afterwards, the effluent pH of AMD permeated sample decreased from pH 12 to pH 8 within 7 pore volumes. It was also noted that effluent pH remained alkaline at pH above 8 till the end of testing for both samples.

In summary, the compacted fly ash samples in the 1<sup>st</sup> and 2<sup>nd</sup> test series maintained an alkaline pH for a considerable period under the AMD permeation, as shown in Figs. 7 and 8. It implied that coal fly ash can generate an alkaline environment, thus immobilize migration of heavy metals.

#### 4.4 Trace elemental concentration

During the permeation tests, concentrations of regulated elements were measured periodically. To illustrate, Fig. 9 presents the concentrations of As and Mo during the 1<sup>st</sup> test series on AFA samples, and Fig. 10 presents concentrations of B and Cr during the 2<sup>nd</sup> test series on NFA samples. It is found that there is no significant concentration difference in the effluent between water and AMD permeated samples for these elements. It was also noted that the concentrations of these elements are far below the leachate criteria set by the regulatory authority (MOE 2000)<sup>11</sup>.

## 5. CONCLUSIONS

The following conclusions can be made based on the preliminary results of laboratory column leaching tests on two coal fly ash samples:

- The break-through hydraulic gradient required for seepage of AMD in coal fly ash is up to 10 times higher than that of water.
- When permeated with AMD (pH 3.8), the compacted coal fly ash samples demonstrated more than two orders of magnitude decrease in the hydraulic conductivity.
- The effluent from compacted coal fly ash samples remained alkaline after more than 12 pore volumes of seepage flow.
- The concentrations of regulated trace elements were well below the leachate criteria set by the local regulatory authority.

## 6. ACKNOWLEDGEMENT

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Fig. 1. Location of Atikokan & Nanticoke coal-fired power plants

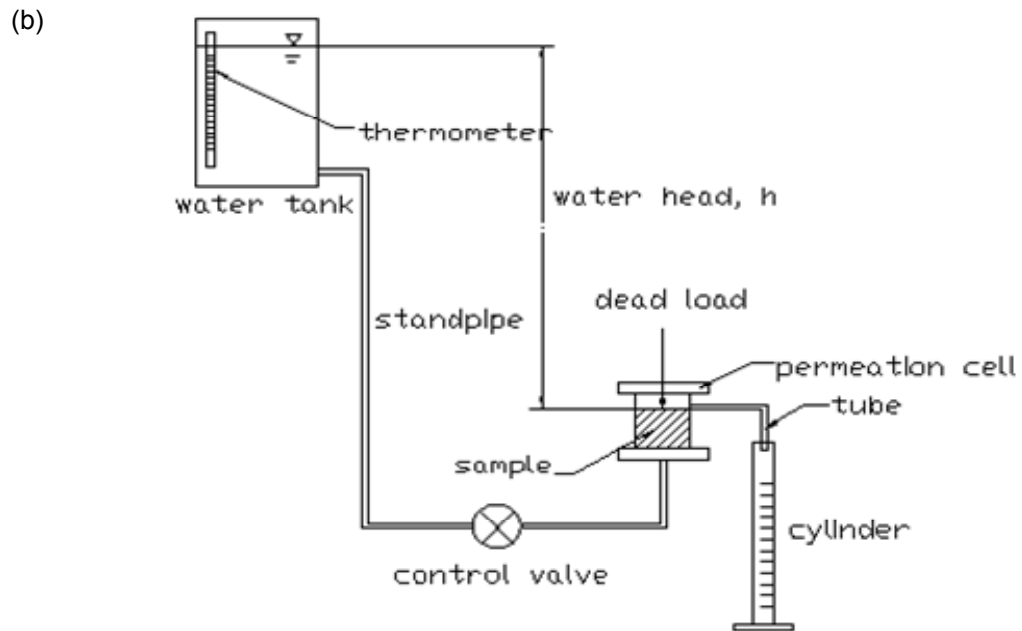
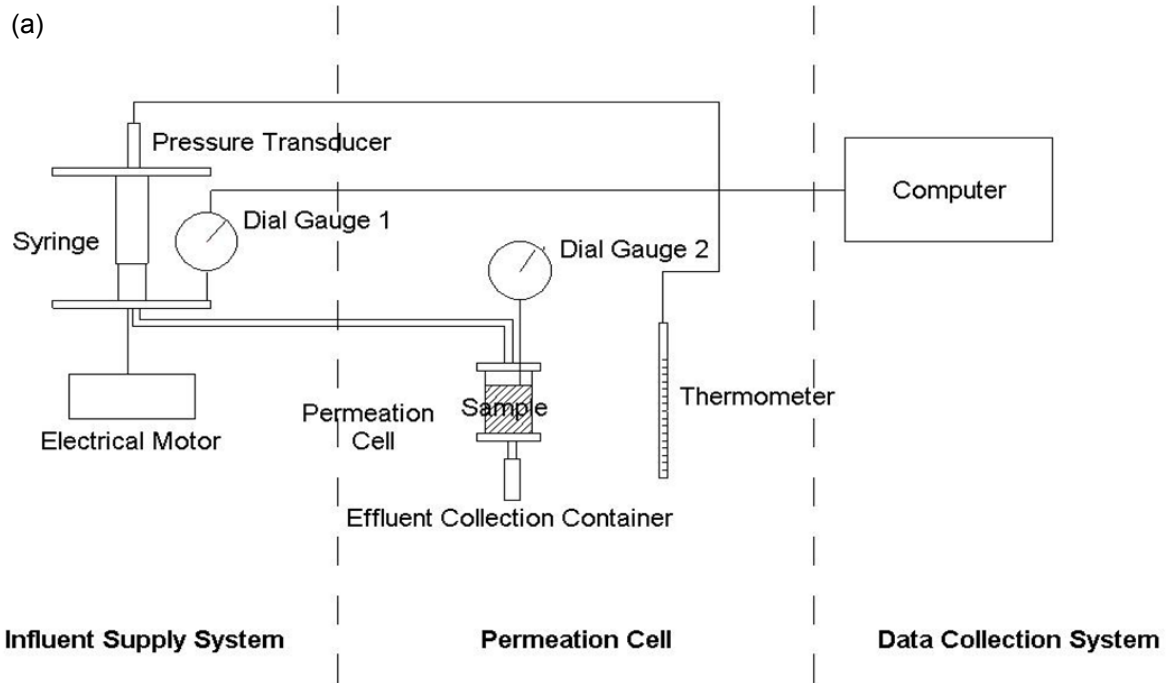


Fig. 2. Schematic diagram of (a) fixed-wall permeator, (b) constant head permeator

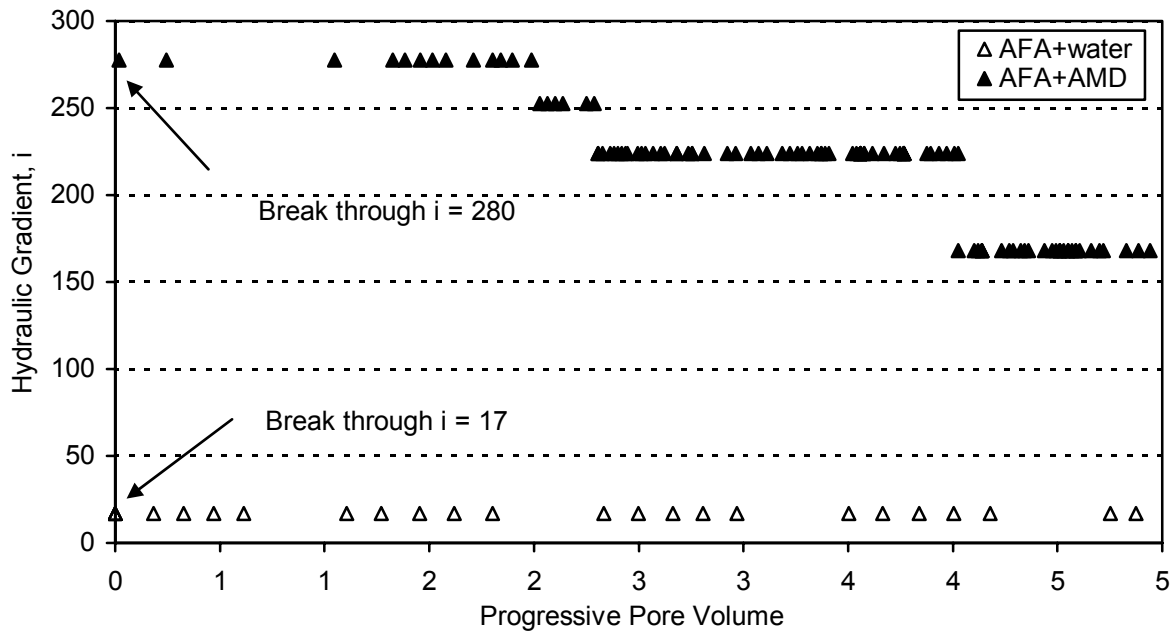


Fig. 3. Hydraulic gradient of Atikokan fly ash permeated by water & AMD

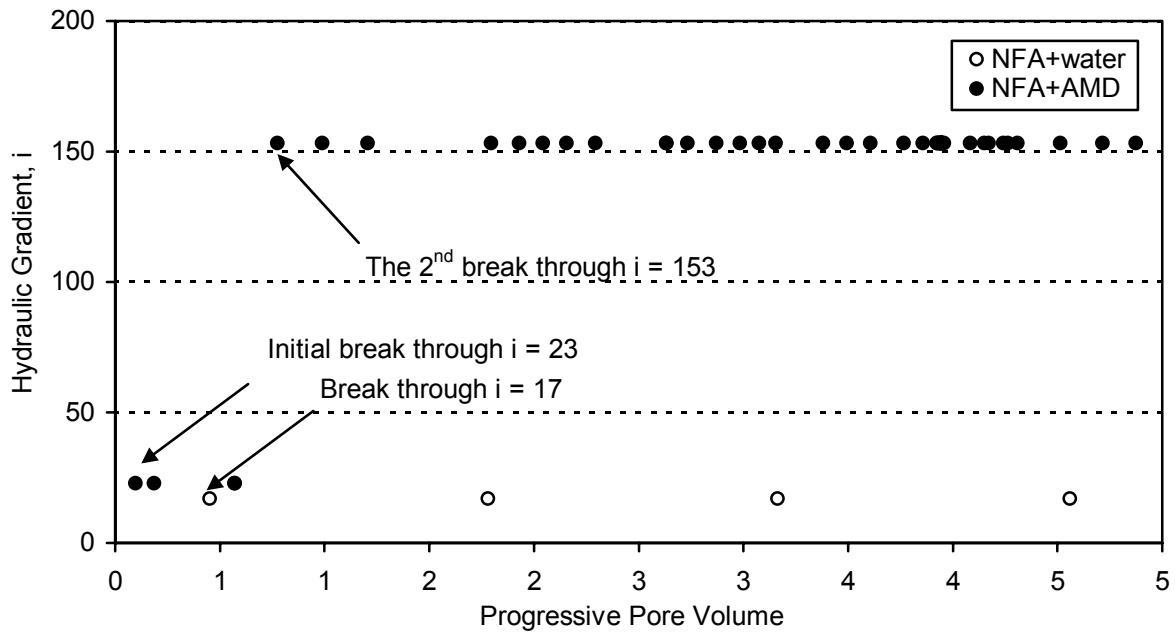


Fig. 4. Hydraulic gradient of Nanticoke fly ash permeated by water & AMD

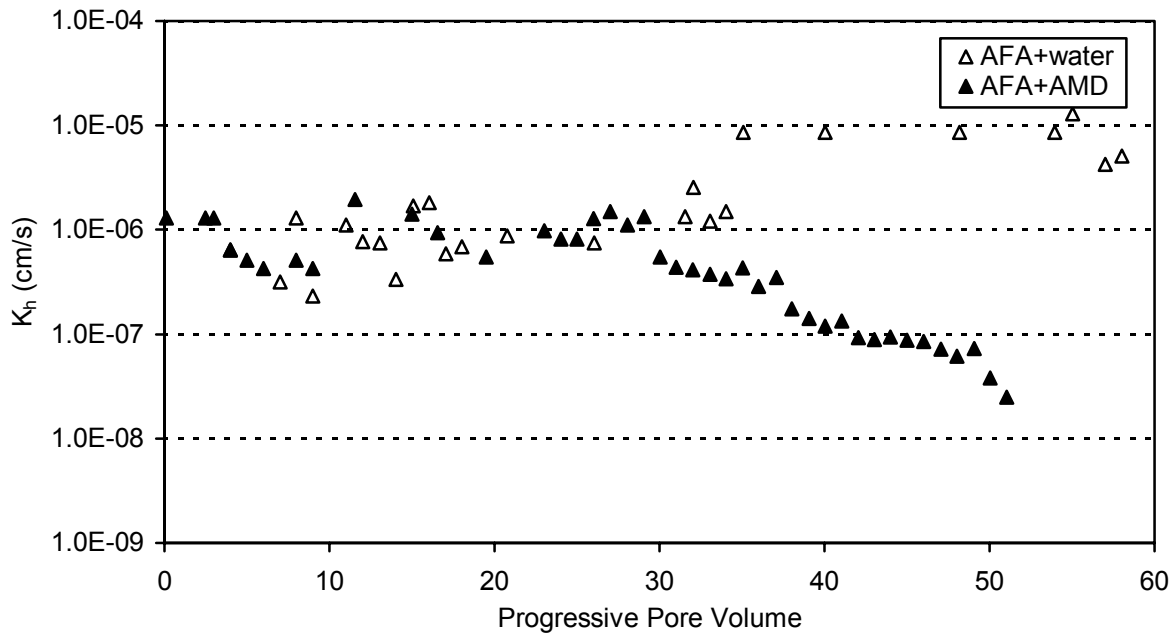


Fig. 5. Hydraulic conductivity of Atikokan fly ash permeated by water & AMD

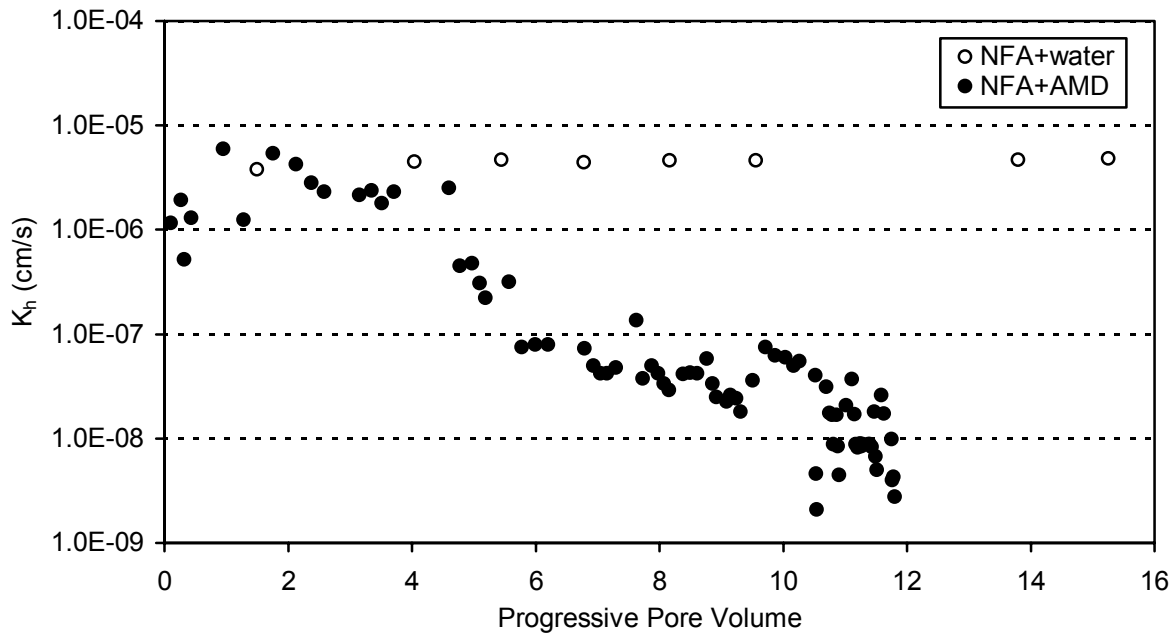


Fig. 6. Hydraulic conductivity of Nanticoke fly ash permeated by water & AMD

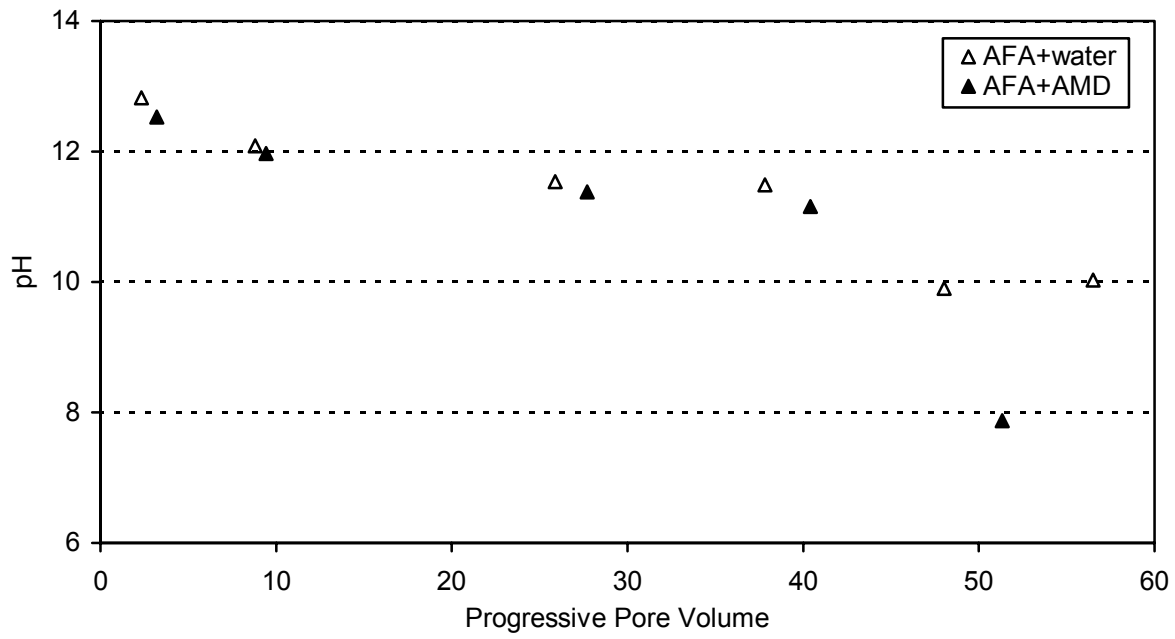


Fig. 7. Effluent pH of Atikokan fly ash permeated by water & AMD

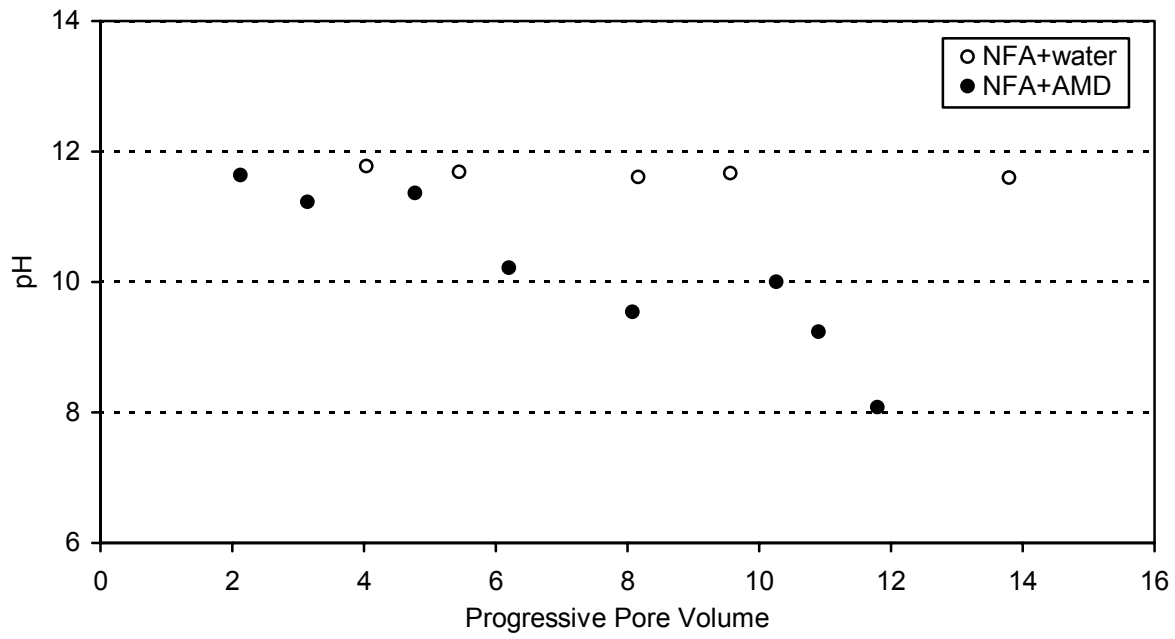


Fig. 8. Effluent pH of Nanticoke fly ash permeated by water & AMD



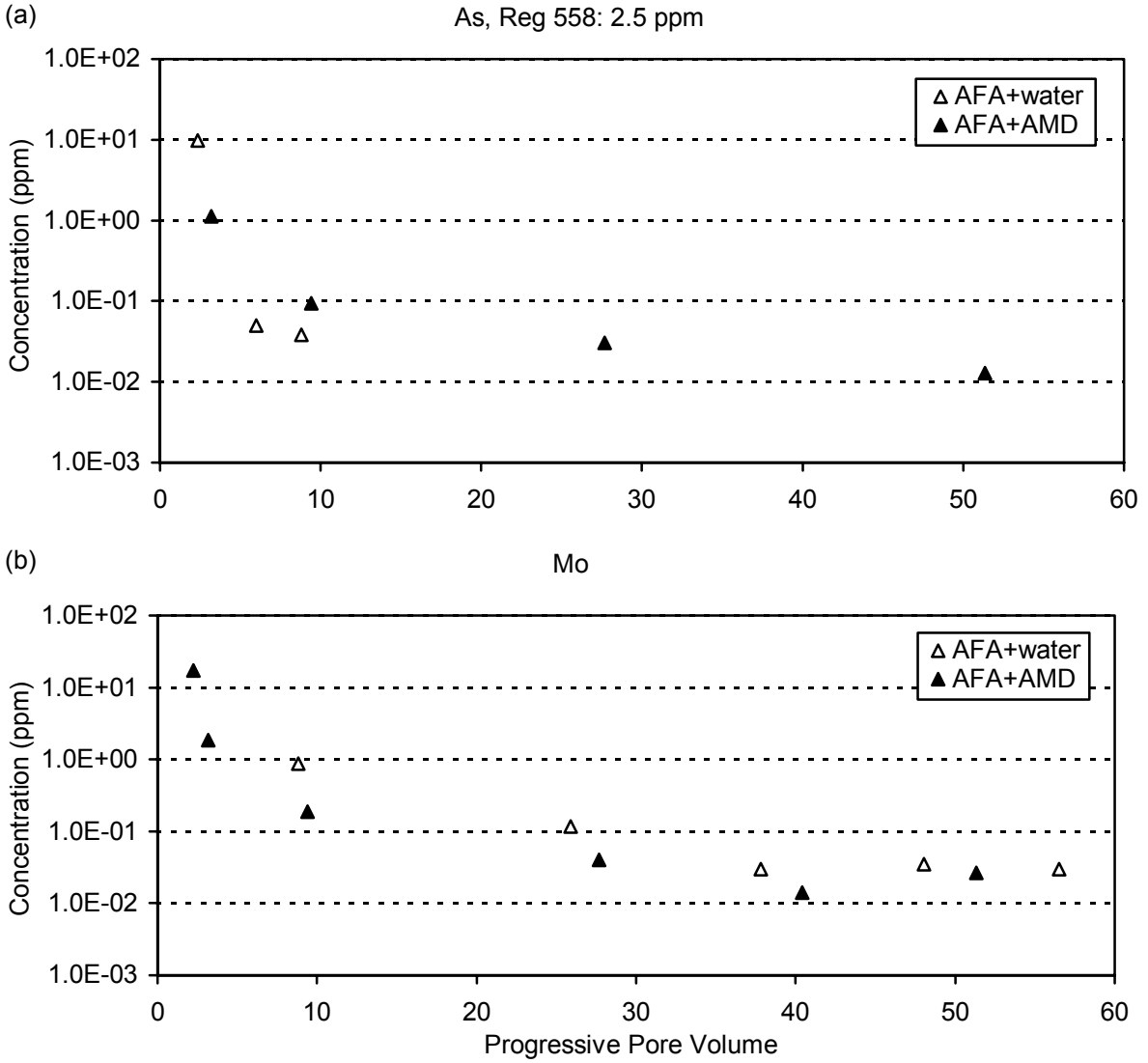


Fig. 9. Concentrations of trace elements (a) As, (b) Mo when Atikokan fly ash permeated by water and AMD

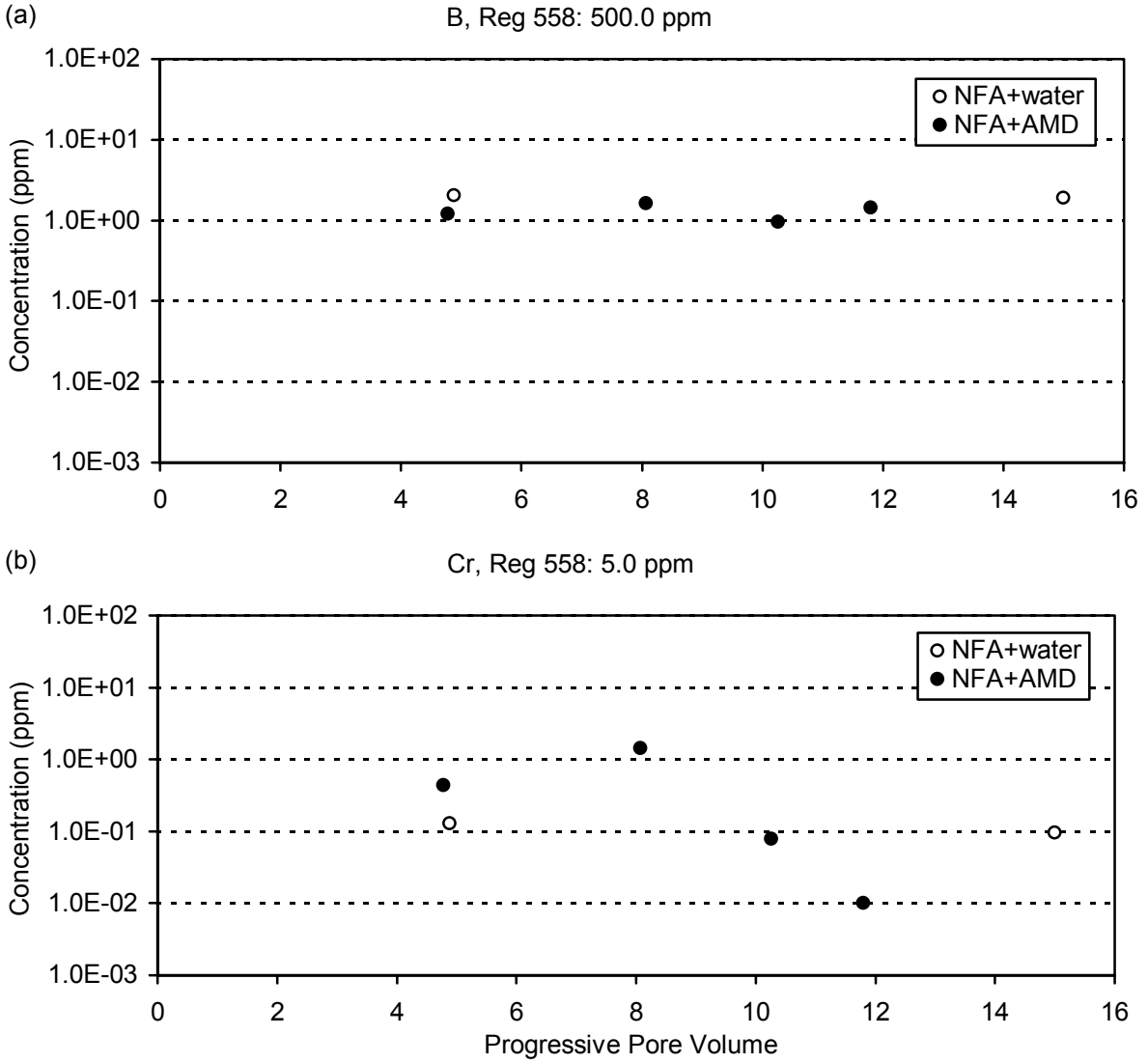


Fig. 10. Concentrations of trace elements (a) B, (b) Cr when Nanticoke fly ash permeated by water and AMD

Table 1. Characterization of Atikokan coal fly ash & Nanticoke coal fly ash

		Atikokan Fly Ash, AFA	Nanticoke Fly Ash, NFA
<b>I. Physical Properties</b>			
Colour		tan	black
Grain Size (USCS)	Sand (4.75-0.075mm)	7.4 %	15.4 %
	Silt (0.075-0.002 mm)	81.6 %	76.1 %
	Clay (<0.002 mm)	11 %	8.5 %
Specific Gravity, G <sub>s</sub>		2.69	2.37
Optimal water content, ω <sub>opt</sub> (%)		11	25.00
Dry Density, ρ <sub>d</sub> (Mg/m <sup>3</sup> )		1.925	1.275
<b>II. Chemical Compositions</b>			
1	SiO <sub>2</sub>	37.99 %	42.62 %
2	TiO <sub>2</sub>	0.68 %	1.27 %
3	Al <sub>2</sub> O <sub>3</sub>	19.92 %	22.68 %
4	Fe <sub>2</sub> O <sub>3</sub>	6.17 %	5.56 %
5	MnO	0.03 %	0.02 %
6	MgO	3.52 %	2.82 %
7	CaO	15.66 %	11.53 %
8	K <sub>2</sub> O	0.62 %	1.17 %
9	Na <sub>2</sub> O	9.3 %	0.52 %
10	P <sub>2</sub> O <sub>5</sub>	0.39 %	0.60 %
11	Cr <sub>2</sub> O <sub>3</sub>	0 %	0.02 %
12	C / LOI	0.84 %	8.06 %
	Total	95.12 %	96.87 %
<b>III. Detected Minerals</b>			
1	Quartz, SiO <sub>2</sub>	X	X
2	Mullite, Al <sub>6</sub> Si <sub>2</sub> O <sub>13</sub>	-	X
<b>IV. Carbonates</b>			
1	Dolomite, CaMg(CO <sub>3</sub> ) <sub>2</sub>	0.71 %	0.00 %
2	Calcite, CaCO <sub>3</sub>	0.58 %	0.82 %

Note:

X - detected minerals

Table 2. Chemical Analysis of AMD

pH	3.77		
$\kappa$	1.98 mS/cm		
<b>Titration Test</b>	EQL (ppm)	MOE regulation 558/00, (ppm)	Concentration, C (ppm)
Cl <sup>-</sup>	1 ~ 5	-	181.504
SO <sub>4</sub> <sup>2-</sup>	3 ~ 4	-	999.700
<b>ICP Test</b>	EQL (ppm)	MOE regulation 558/00, (ppm)	Concentration, C (ppm)
Aluminum Al	0.010	-	10.900
Arsenic As	0.010	2.5	0.061
Barium Ba	0.010	100.0	0.015
Beryllium Be	0.010	-	0.001
Boron B	0.010	500.0	0.051
Cadmium Cd	0.010	0.5	0.006
Calcium Ca	0.010	-	242.000
Chromium Cr	0.010	5.0	0.000
Cobalt Co	0.010	-	0.540
Copper Cu	0.010	-	3.120
Iron Fe	0.010	-	0.090
Lead Pb	0.010	5.0	0.040
Magnesium Mg	0.010	-	38.000
Manganese Mn	0.010	-	1.840
Molybdenum Mo	0.010	-	0.000
Nickel Ni	0.010	-	21.100
Potassium K	0.010	-	20.600
Silver Ag	0.010	5	0.002
Sodium Na	0.010	-	95.400
Vanadium V	0.010	-	0.012
Zinc Zn	0.010	-	0.739
Selenium Se	0.010	1.0	0.000
Sulphur S	0.010	-	250.000

Note:

EQL - Estimated quantitation limit