

# Evaluating the Interaction of Coal Ash Leachates with Rock Materials for Mine Backfill Studies

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## ABSTRACT

Two different laboratory-based leaching test protocols have been developed to evaluate the chemical interaction between ash, water and different mine-site rock materials, as a basis for assessing the potential environmental impact of using coal combustion products as backfill in mining operations. One was a two-stage serial batch testing program in which leachates from representative ash samples were allowed to interact separately with relevant rock materials under laboratory conditions; this was intended to duplicate the reactions expected when ash leachate percolates into surrounding rock strata. The other, intended to evaluate the leachate characteristics expected from interaction of water with an intimate admixture of ash and rock materials was a single-stage mixed batch testing program where mixtures of the ash and relevant rock samples were extracted with water.

The concentrations of major and trace elements released from both tests were compared to the concentrations expected from the leaching characteristics of the individual ash and rock samples when tested alone. Lower than expected concentrations of several environmentally significant elements were found in solution after interaction of the ash leachate with the rock samples. This indicated that the elements released from the ash in a mine backfill may be significantly attenuated when the ash leachate comes into contact with other rock materials. The extent of attenuation was greatest for those rocks with high proportions of clay minerals (e.g. shales) or iron oxy-hydroxides (e.g. iron-stained sandstones). Rocks such as white quartz sandstone, with lower clay or iron mineral contents, appear to have a lesser attenuating effect.

## INTRODUCTION

### *Use of ash in mine backfill*

Backfill of mine excavations with ash is gaining increased attention from the power and mining industries as a more beneficial process of ash disposal<sup>1,2,3</sup>. The main beneficial use of ash for mine backfill has traditionally been derived from the interaction of alkaline

ash with mine solids, mine waters or in mining voids to ameliorate acid mine drainage (AMD) conditions<sup>4,5</sup>. However, ash is also routinely emplaced in open-cut mines as part of void infill programs, such as in the western USA<sup>6,7</sup>, without necessarily an AMD involvement.

In underground mines ash-based backfill may be used for ground support and subsidence control, an area in which the critical factors are flowability, density, porosity, abrasiveness, strength and pozzolanic properties<sup>8</sup>. Fly ash has also been used for the control of mine fires<sup>9,10</sup>, as a contaminant barrier to reduce the escape of waterborne contaminants from mine products such as preparation tailings<sup>11,12</sup>, and as an additive to enhance the fertility of mine soils in reclamation programs<sup>13,14</sup>.

### ***Environmental evaluation of ash for backfill applications***

A considerable amount of research has been carried out on the interaction of coal ash with water, with special focus on the mobility of environmentally significant trace elements in short and long-term leaching processes<sup>15,16,17</sup>. The pH of the ash-water system is a major control on the mobility of different elements<sup>18,19</sup>. This in turn depends on the chemistry of the ash, particularly the balance between the concentration of alkaline-earth elements, Ca and Mg on the one hand and the proportion of potentially acid-generating SO<sub>3</sub> and P<sub>2</sub>O<sub>5</sub> on the other<sup>20</sup>. Australian ashes may range from fairly acid (around pH = 4) to relatively alkaline (pH = 12), depending on the ash chemistry<sup>18</sup>; the chemistry of the ash, especially the glass fraction, is in turn controlled by the mineral matter in the feed coal<sup>21</sup>.

Most research on ash leaching has been focused on two-component ash-water systems. The interactions when ash used as mine backfill, however, occur within three-component systems involving the ash, the mine water or groundwater, and the rock strata in and around the mine site. Programs to establish the environmental behaviour of a particular ash in a given mine backfill application should therefore include studies not only of element mobilities when the ash itself is exposed to water under the conditions (pH, Eh etc.) expected at the site, but also the interaction of the leachate from the ash and the mine water with the associated rock strata. Examples of the latter process include ion exchange reactions with clay minerals, solution of carbonates, and precipitation of salts such as sulphates, carbonates or silicate materials.

A logical extension of the two-component approach is therefore to investigate the more complex ash-water-rock system in a mine-site context. Although a significant amount of work has been carried out on the interaction of alkaline ash leachates with mine rocks in situations associated with acid mine drainage, only limited information is available on assessment methods and protocols for assessing the interactions between ash, rock and water in other situations (e.g. acid ashes and/or mines without AMD), as a basis for better managing any potential environmental risks associated with more generic mine backfill operations.

## SAMPLES STUDIED

To test the framework for evaluating more fully the possible processes involved, a series of studies was undertaken to investigate the interactions between the fly ash from a number of different Australian power stations and a range of rock types from nearby open-cut coal mines. Most of the fly ash at these power stations is currently emplaced in ash ponds. However, the ponds are approaching the limits of their storage capacity, and other sites, including worked-out areas of adjacent open-cut coal mines, are therefore under consideration for future ash emplacement. New mining and power station projects in these and other areas are also considering ash placement in mines as the principal basis of future ash management programs.

Fresh fly ash samples were obtained from the Tarong North Power Station in the Tarong Basin of Queensland and the Collie and Muja Power Stations in the Collie Basin of Western Australia. A range of rock samples was taken from open-cut coal mines associated with the power stations, representing the range of materials with which leachates from the ash might come into contact in any backfill operations. These included different types of sandstone, shale and, in one case, coal preparation refuse (Table 1).

**Table 1:** Ash and rock samples evaluated for the study

| <b>Sample Details</b>            | <b>Abbreviation</b> | <b>pH</b> | <b>EC</b> |
|----------------------------------|---------------------|-----------|-----------|
| Tarong North dense phase ash     | Ash                 | 6.97      | 488       |
| Meandu Mine white sandstone      | White s/s           | 6.49      | 71        |
| Meandu Mine shale                | Shale               | 6.45      | 137       |
| Meandu washery tailings          | Tailings            | 6.22      | 205       |
| Collie fly ash                   | Collie              | 4.13      | 2470      |
| Ewington Mine light grey shale   | Lt gy sh            | 4.85      | 80        |
| Ewington Mine hard sandstone     | Hd ss               | 3.54      | 554       |
| Ewington Mine white sandstone    | Wh ss               | 5.60      | 37        |
| Ewington Mine carbonaceous shale | Carb sh             | 4.38      | 169       |
| Muja C-D fly ash                 | Muja                | 5.50      | 1665      |
| Premier Mine white sandstone     | Wh ss               | 6.04      | 37        |
| Premier Mine grey shale          | Gy sh               | 4.40      | 342       |

**Note:** pH and EC ( $\mu\text{S}\cdot\text{cm}^{-1}$ ) taken from 3-day single batch tests

The rock samples were crushed to <5 mm, and representative splits further ground to <63  $\mu\text{m}$  for chemical and mineralogical analysis. The ashes were dried where necessary, and representative splits also finely ground for chemical and mineralogical studies. A range of analytical techniques was used to characterise the materials, including X-ray fluorescence spectrometry, X-ray powder diffraction, ICP-AES and ICP-MS techniques.

## LEACHING CHARACTERISTICS OF ASH AND ROCK SAMPLES

An initial series of leaching tests (referred to as individual batch tests) were carried out on each ash (dried but otherwise as received) and each rock sample (dried and crushed to <5 mm) over three different time periods (1, 3 and 7 days) under otherwise identical conditions<sup>18</sup>. Although it is preferable that such testing should use groundwater from the mine site<sup>16</sup> to allow for the impact of its natural pH or any elements already in solution, this was impractical for the present study due to the distant location of the test site with respect to the laboratory and the possible loss of some ions (e.g.  $\text{HCO}_3^-$ ) from the water with transport and storage. De-ionised water was therefore used as the leaching medium for the laboratory leaching test program.

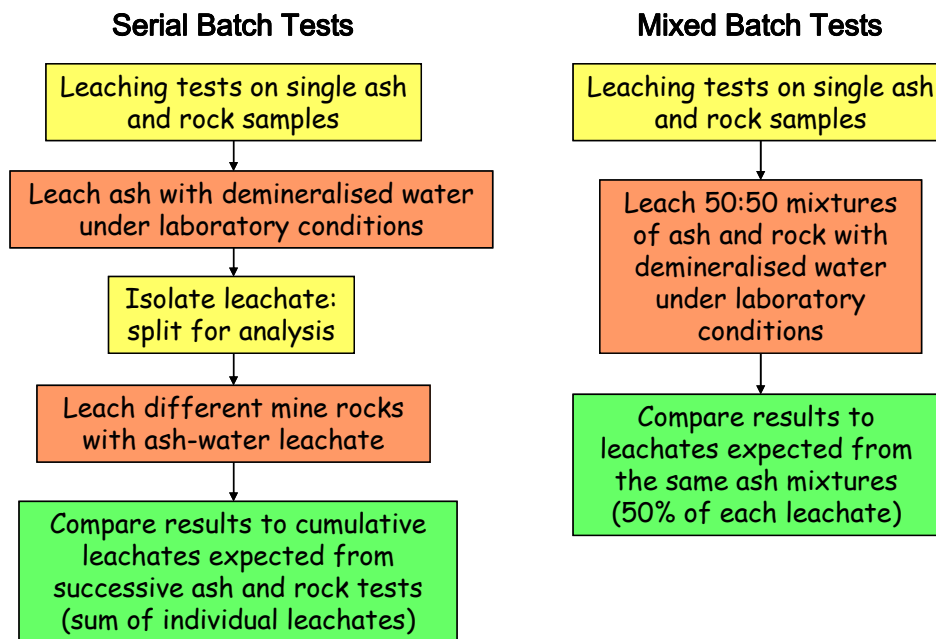
Samples of 32 grams of ash or crushed rock were weighed into polyurethane bottles and 112 grams of deionised water added, making the liquid:solid ratio 3.5:1. The bottles were sealed and shaken on a horizontal shaker at 110 rpm at room temperature for the required time. After the required shaking time the solutions in each bottle were separated from the solid phase by drawing through 0.22  $\mu\text{m}$  cellulose acetate membrane filters. Approximately 30 ml of each extract was immediately acidified using 10 drops of concentrated nitric acid prior to major and trace element determinations using ICP-AES and ICP-MS techniques. The remaining non-acidified leachates were subjected immediately after sample collection to measurement of pH and electrical conductivity (EC), and to  $\text{HCO}_3^-$  determination by titration.

## EVALUATION OF ASH-ROCK-WATER INTERACTIONS

The individual batch tests provided data on the behaviour of the ash and rock samples when exposed individually to water. In order to assess the possible interactions between the ash leachates and relevant rock strata in a mine backfill situation, however, a specially developed series of serial batch tests and mixed-batch leaching tests was then used (Figure 1), in order to assess how the ashes might be expected to behave in a three-component ash-rock-water system.

### ***Serial Batch Tests***

Serial batch tests involve leaching of each ash sample and testing the interaction of the leachate from that ash with samples of rock materials from the relevant mine site (Table 1). These tests are intended to evaluate the impact of a scenario in which water passes through an ash-filled mine void and then into different types of adjacent rock materials. In such a scenario, the water would first reach equilibrium with the ash, after which a second equilibrium would be developed between the ash leachate and the surrounding rock strata.



**Figure 1:** Flow-sheet illustrating two-stage serial batch tests (left) and 50:50 mixed batch tests (right) used in the mine backfill study.

Six samples of 32 grams of each ash were weighed into polyurethane bottles and 112 grams of deionised water added, making the liquid:solid ratio 3.5:1 as for the single-stage tests on the same material. The bottles were sealed and shaken on a horizontal shaker at 110 rpm at room temperature for 3 days, after which the solutions in each bottle were separated from the solid phase by filtration through 0.22  $\mu\text{m}$  cellulose acetate membrane filters. Leachates from all six samples were pooled for each ash. Approximately 30 ml of the leachate was immediately acidified, after which it was analysed by ICP-AES and ICP-MS techniques, as a check against the individual batch test results for the same ash sample. The pH and EC of the individual ash leachates were also re-determined.

Samples of 32 grams of each rock were weighed into polyurethane bottles and 112 grams of the remaining relevant ash leachate added, making the liquid:solid ratio again 3.5:1. The bottles were sealed and shaken on a horizontal shaker at 110 rpm at room temperature for 7 days, after which the solutions in each bottle were filtered, aliquots acidified for analysis of their major and trace components, and other aliquots subjected to pH and EC determination.

### **Mixed Batch Tests**

Mixed batch tests, involve leaching experiments on mixtures made up of each ash and each of the relevant rock samples. These were intended to represent a situation in which ash and rock were in contact with the water simultaneously, such as might occur where a blend of ash and mine rock is used as the backfill material. The equilibrium developed in this case would not necessarily be the same as with the serial tests; the

ash would remain in contact with the water in such cases and a three-way equilibrium involving ash, rock and water would be developed.

Samples of 32 grams containing 50:50 mixtures of each ash and each of the relevant rock samples were weighed into polyurethane bottles for the mixed-batch tests, and 112 grams of deionised water added to make the liquid:solid ratio 3.5:1. The bottles were sealed and shaken on a horizontal shaker at 110 rpm at room temperature for 7 days. The solutions in each bottle were then filtered, aliquots acidified, and the acidified and untreated samples analysed as for the serial batch tests.

## **SUMMARY OF TEST RESULTS**

Plots showing the results obtained for pH, electrical conductivity, and selected major and trace elements in each sample for the serial batch and mixed batch tests are presented in Figures 2 and 3. The graphs show the concentrations obtained in comparison to the expected values in each case, derived from the individual batch test results as described in more detail below. Note that the concentration values on the vertical axis of each plot are different, reflecting the absolute values of concentration in solution for each of the elements and samples involved.

### ***Serial Batch Tests***

To provide a basis for evaluating the results from the serial leaching tests, the concentration of each element (and also the pH and electrical conductivity) expected after the two stages of each serial batch test was taken as the sum of the concentrations of that element released from the ash after three days in the single-batch test and the rock in question after seven days in the respective single-batch test. Such an expectation is based on the assumption that the elements released from each material in the two-stage test would be additive, i.e. if a concentration of  $x$  units was released from the ash after the first stage of the experiment and a concentration of  $y$  units was added to the leachate from the rock sample in the second stage of the test, the expected element concentration from the two-stage process would be  $x+y$  units.

The results obtained from the serial batch tests for selected components in relation to these expectations, as well as for the same components released from each individual ash itself in two separate individual batch experiments, are summarised in Figure 2.

The validity of the assumptions in relation to the overall expectations appears to be borne out by the results obtained from the electrical conductivity data for the single-batch (Table 1) and serial tests. Although each ash sample when tested alone had a relatively high conductivity in relation to the rocks in the relevant test series, the conductivity of the leachates from most of the serial tests was higher still, with the increase in each case being very close to the conductivity of the relevant rock sample when tested individually in the single-stage study.

Although there is some variation, the pH values of the leachates after the tests were in most cases close to the levels expected from single batch tests of the respective ash and rock samples. This suggests that these rocks have a significant buffering capacity, probably derived from their abundant clay minerals. In other cases, especially with the sandstones, the pH remained similar to that of the relevant ash leachate found in the single batch tests, suggesting that the rock in question has a relatively low buffering capacity, possibly because of a lower clay mineral content.

The concentration of the various elements in solution when ash leachates are exposed to different rock types may be expected to fall into one of three categories:

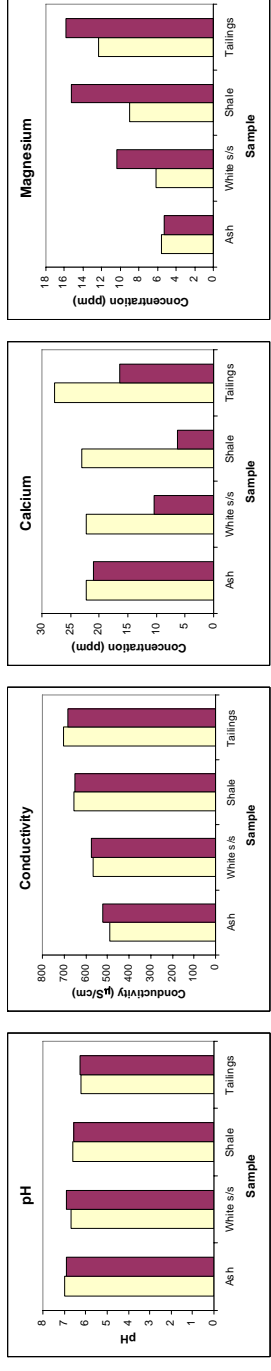
- a) The element concentration in the leachate increases to a level approximating that expected from the combination of the two individual leachate solutions, suggesting that the rock in question continues to release a particular concentration of that element regardless of the other components (in this case derived from the ash) also present in the surrounding solution;
- b) The element concentration increases above the expected level, suggesting that the rock releases a greater concentration of that element when the ash leachate is introduced than it would with water alone;
- c) The element concentration is less than that expected from the combined leachates, suggesting that the presence of the rock material causes no more, and indeed possibly less of the element to be released from the rock when the ash leachate is present. Two possibilities arise in such circumstances:
  - i. The concentration associated with the rock remains essentially the same as with the ash alone, suggesting either that the element is at saturation with respect to the rock, or that the rock has no impact on the element concentration released by the ash sample. This is equivalent to option (a) above, in cases where the rock when tested alone releases no significant concentration of the element in question.
  - ii. The element concentration after interacting with the rock falls significantly below that in the original ash leachate, suggesting that the element in question may have either been precipitated in some form or it may have been taken up by some other process into the rock components (e.g. by ion exchange, absorption, adsorption).

Calcium and aluminium for the Collie Basin samples and sulphur (expressed as sulphate) for all of the ashes are the only elements among those plotted in Figure 2 where the concentrations in the second leachate were consistently close to those expected by addition of the individual leachate solutions (category (a) above). A number of other elements that were not plotted, such as silicon, sodium and boron, also show this type of behaviour<sup>22, 23</sup>.

### Tarong

#### Serial Batch Tests

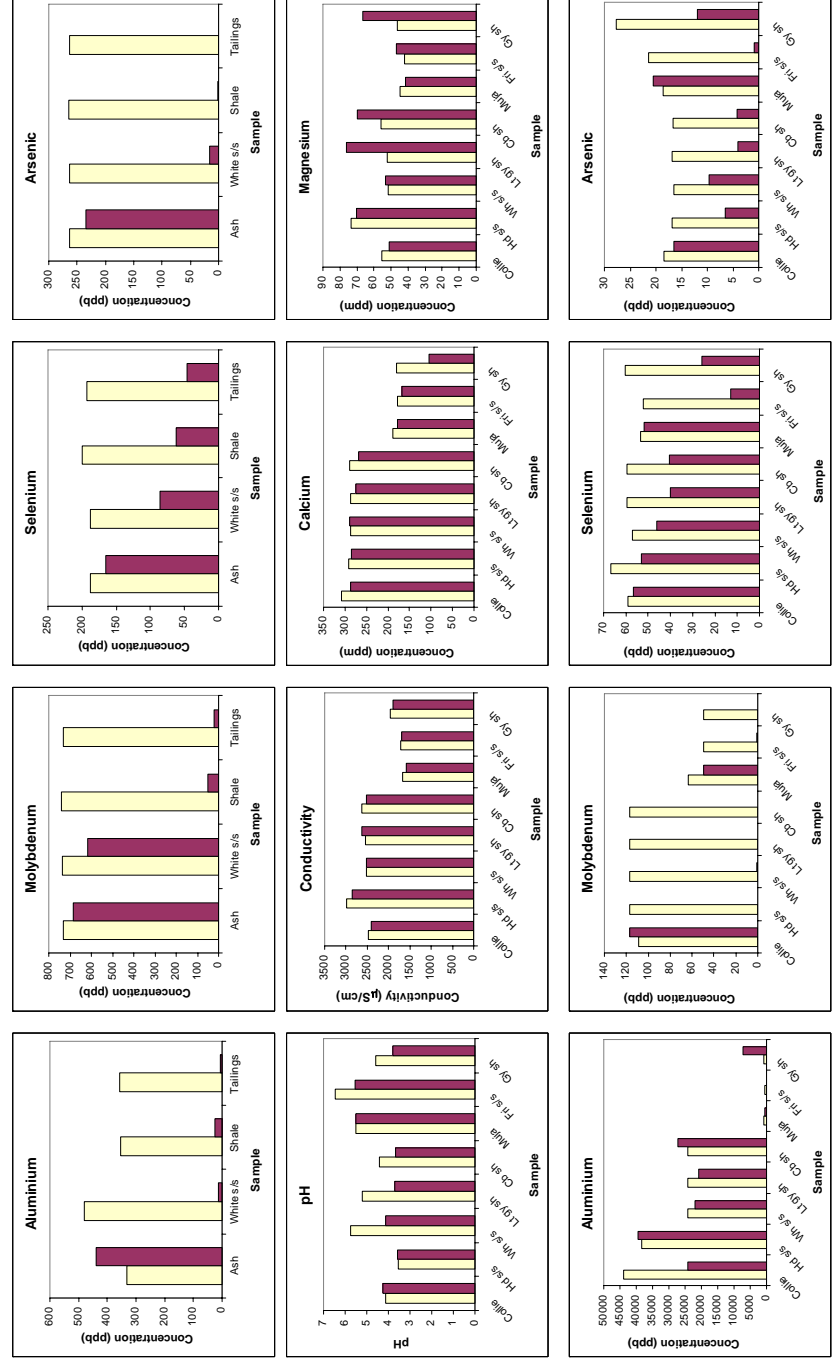
Expected █  
Observed █



### Collie and Muja

#### Serial Batch Tests

Expected █  
Observed █



**Figure 2:** Data for pH, EC and concentration of Ca, Mg, SO<sub>4</sub>, Al, Mo, Se and As in solution from serial batch tests of Tarong (top) and Collie and Muja (bottom) ash leachates on relevant rock samples.

Magnesium shows a higher concentration than expected in the leachates (category (b) above) for all of the Tarong rock samples and for the shale samples from the Collie Basin, suggesting that additional release was stimulated by introduction of the ash leachate solution. The Mg concentrations from the sandstones tested with the Collie Basin ashes, however, were close to the levels expected from the solution mixing calculations.

Most of the remaining elements, however, including Ca and Al from the Tarong samples and Mo, Se and As from the entire sample series, although often relatively mobile from the ash alone, have much lower concentrations after the ash leachate has interacted with the rock samples (category (c)ii). The sandstone samples in some of these cases (e.g. Mo for Tarong) do not seem to have had a great impact (category (c)i), but the concentrations in solution are significantly lower after exposure to the shale and the coal tailings materials from Tarong as well as most of the rocks in the Collie Basin study.

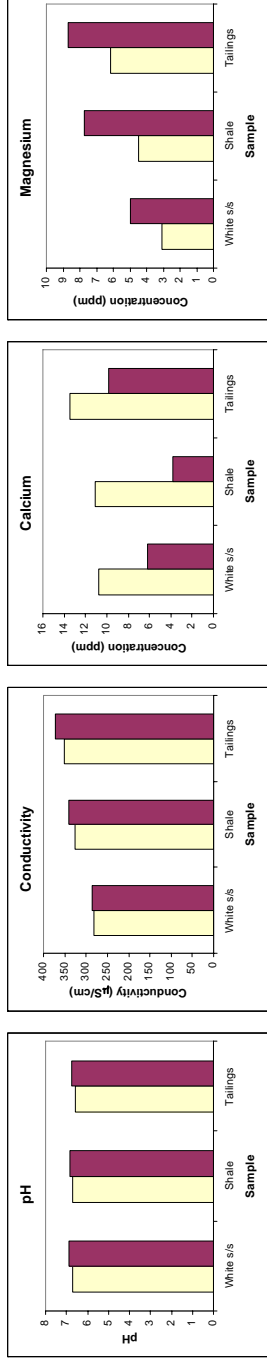
### ***Mixed Batch Tests***

To provide a basis for evaluation of the results from the mixed batch tests, the concentration of each element (and also the pH and electrical conductivity) expected after leaching of the ash-rock mixture was estimated based on half the sum of the concentrations of that element released from the ash after three days and the rock in question after seven days in the single-batch tests. Such an expectation is based on the fact that only half the mass of ash and half the mass of rock was in contact with the volume of leaching solution used, compared to each of the relevant single-batch tests. Hence, the elements released from each material in the mixed-batch test, if additive, would be the sum of those two half-concentrations: i.e. if a concentration of  $x$  units was released into the test volume from the ash in the relevant single sample test and a concentration of  $y$  units was released into that volume from the rock sample in the relevant single sample test, the expected element concentration from the mixed batch process would be  $(x+y)/2$  units.

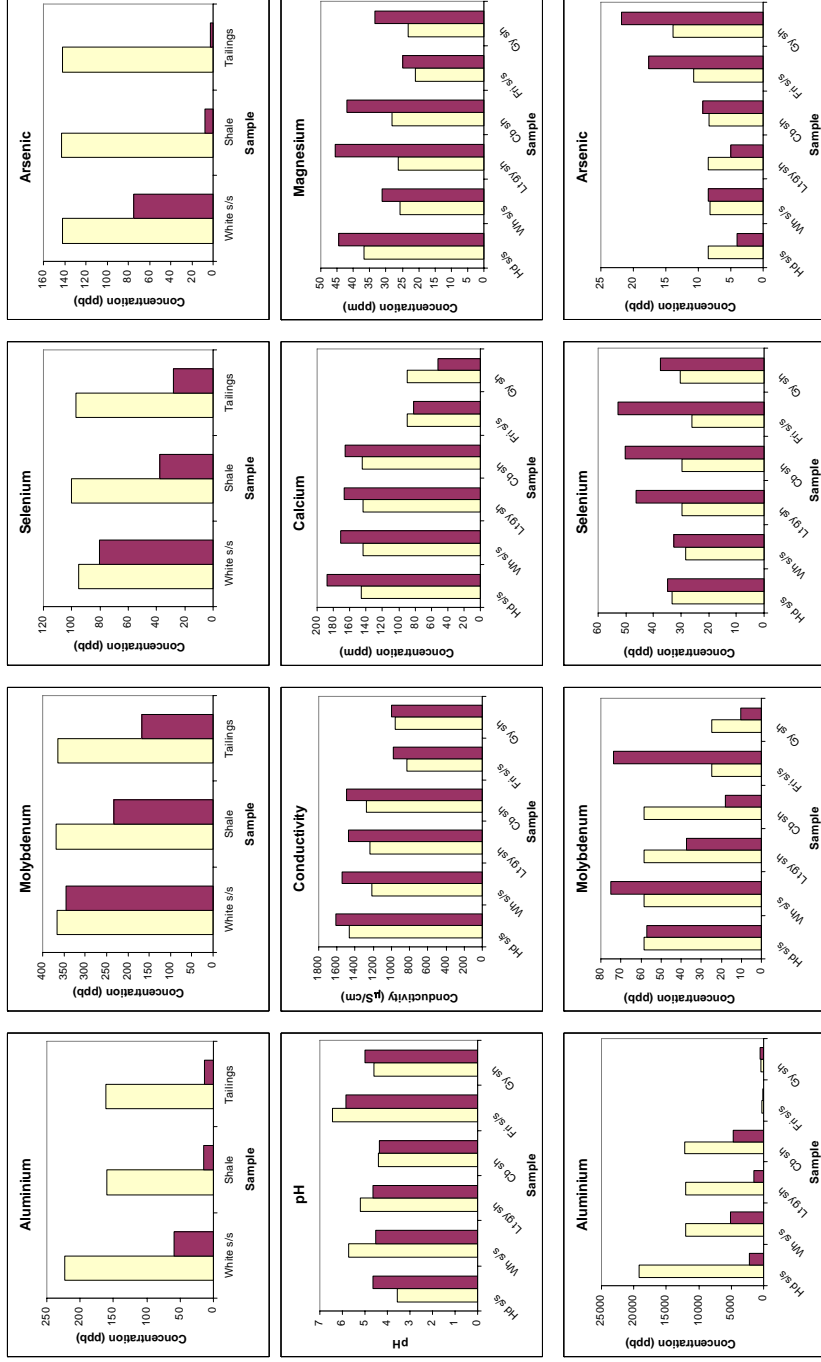
The results for the mixed-batch leaching tests (Figure 3) are similar overall to those of the serial leaching tests, especially for the Tarong sample series, but with lesser differences between ash and ash plus rock than with the two-stage serial test program. The leachate pH values for the ash-rock mixtures are relatively close to expectations, but most tend to be closer to the pH of the leachate from the ash with water alone (Table 1) than to the pH of the respective rock materials. This suggests that the buffering capacity of the ash is stronger than that of the rock samples, and hence that the pH of an ash-rock mixture, at least for the samples studied, would probably depend more on the ash characteristics than on the rock type involved.

The electrical conductivities of the mixed-batch leachates were close to but generally slightly higher than the values expected based on the individual leaching tests. As with the serial batch tests, this suggests that elements released from both components contribute to the overall leachate derived from the test procedure. The individual elements, however, behave somewhat differently, as indicated in the discussion below.

**Tarong**  
**Mixed Batch Tests**



**Collie and Muja**  
**Mixed Batch Tests**



**Figure 3: Data for pH, EC and concentration of Ca, Mg, SO<sub>4</sub>, Al, Mo, Se and As in solution from mixed batch tests of Tarong (top) and Collie and Muja (bottom) ash leachates on relevant rock samples.**

As with the serial batch tests, the concentrations of sulphur (as SO<sub>4</sub>) for all samples and Ca for the Collie Basin materials was close to expectations in the mixed batch tests (Figure 3). In contrast to the serial test results, however, the concentration of Al in solution with the tests involving the ash from Collie Power Station was considerably below expectations, displaying a similar pattern to the Tarong ash and rock samples.

In addition to Al as discussed above, the concentration of Mo was also lower than expectations for the tests in the mixed batch series involving shales and coal tailings, but close to or above expectations when the ashes from both basins were mixed with the respective sandstone samples. The concentrations of Se and As were also lower than expected for the Tarong ash-rock mixtures, but close to or above expectations for the Collie Basin samples.

For those elements where the concentrations in the mixed batch tests were lower than expected it appears, as with the serial tests, that some of the element in question has been taken up by the rock components. Especially with Tarong, but also with Mo in the Collie Basin tests, the effect is more marked in the mixtures involving the shale and tailings samples; sandstones appear to have had less of an impact on the element concentration in the mixed-batch leachate solutions.

## **CONCLUSIONS**

This study has shown that the coal mine rock materials, particularly the overburden shales, clay-rich coal preparation refuse and ferruginous sandstones, may interact with leachates from emplaced ash and reduce the concentration of environmentally significant trace elements that might otherwise be released from that ash in association with an on-site mine backfilling program. This suggests that the concentration of many potentially significant trace elements released into percolating waters from a mine-site ash deposit may be attenuated by contact of the ash leachate with the adjacent rock strata. These properties appear to be related to the nature of the rock in question, and provide a basis for more fully assessing the potential impacts of different ashes in relation to relevant rock strata for mine backfill projects.

The serial batch test procedure provides a more definitive basis for identification of the attenuation effects of different rock materials on a given ash leachate than the mixed batch test procedure. The mixed batch test procedure is a complementary routine that provides an indication of the leachate produced where ash and rock are intimately mixed with each other in the backfill area. Although the most appropriate test program for any individual site would probably depend on the design of the proposed mine backfill operations, collection and evaluation of data from both procedures, in conjunction with regional hydrogeology and other site investigations, may be of value in assessing the environmental impact of particular backfill options.

The exact nature of the interactions identified by the study has not yet been fully determined. However, since the pH of the relevant solutions remains fairly constant it is thought that major precipitation processes associated with pH change are unlikely to be

involved. Because the greatest attenuation effects are associated with rocks rich in clay minerals or poorly crystalline iron oxy-hydroxides, adsorption by the clay minerals or iron oxy-hydroxides in the rocks are thought to be the principal processes responsible for the reduction in trace element concentration when the ash leachate comes into contact with the rock materials. The decrease in concentration of some ions, such as Ca in the Tarong series, and the associated increase in Mg, suggests that in some cases ion exchange involving the clay minerals may also be involved.

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