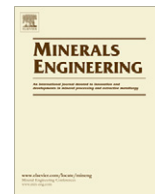




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Effect of fly ash characteristics on the behaviour of pastes prepared under varied brine conditions

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ABSTRACT

To meet the escalating demand of energy more coal ash and brines are inevitably produced as by-products. Large volumes of these wastes and increasing environmental awareness necessitate the development of more sustainable methods to mitigate the environmental footprint. Paste backfill is one of the potential solutions to keep the energy industry sustainable. The behaviour of pastes was investigated by strategically varying brine composition mixed with the two types of fly ash. The results showed that fly ash plays a more prominent role in the behaviour of pastes than brines. It is therefore imperative to consider both fly ash and brine characteristics i.e. constituents of paste for the development of an environmentally sound paste backfill practice. Technically there are numerous benefits in pursuing the proposed solution.

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1. Introduction

The ever-increasing energy demands necessitate combustion of more coal, which is the reliable source of energy worldwide. It is a fact that combustion of low-grade coal generates vast quantities of fly ash of which the global average utilisation is approximately 16% (Ahmaruzzaman, 2010). This leaves the remaining 420 million tons of fly ash requiring measures for disposal annually.

The scarcity of potable water as well as high water consumption during mineral and coal processing leads to the inevitable saline brine production. The management of industrial brines resulting from water recovery processes presents an environmental concern especially inland (Nassar et al., 2008; Souilah et al., 2004; Vedavyasan, 2001), where the option of oceanic disposal is often uneconomical (Ahmed et al., 2003; Nassar et al., 2008; Korngold et al., 2009). The variability of brines as well as failure to meet legal environmental requirements restricts their potential utilisation in applications such as crystallisation of marketable salts, and mixing water in concrete. Literature focuses on the detrimental impact of chlorides in reinforced concrete (Balonis et al., 2010; Neithalath and Jain, 2010; Arya et al., 1990; Barberon et al., 2005) and sulphates to explain concrete deterioration (Medvešček et al., 2006; Collepardi, 2003; Borsoi et al., 2009). These anions give rise to durability problems if they come from

the external environment such as the interaction of seawater with concrete.

The study to utilise seawater as mixing water in concrete produced stronger concrete than a control prepared with potable water (Akinkulore et al., 2007; Taylor and Kuwairi, 1978). Mahlaba and Pretorius (2006) and Mahlaba (2007) indicated that, compared to water, brines have an advantageous effect on the workability of fly ash pastes. The major components in seawater are chloride, sodium, calcium and sulphate (Alahmad, 2010) which most saline brines have been reported to emulate (Ahmed et al., 2003; Ravizky and Nadav, 2007; Mooketsi et al., 2007; Koch, 2002). Furthermore, the use of Cl-bearing compounds to accelerate strength development and improve mechanical properties is common practice in concrete production (Akinkulore et al., 2007; Taylor and Kuwairi, 1978; Shi, 1996). Na₂SO₄ is used to accelerate pozzolanic reactions whereas gypsum (CaSO₄·2H₂O) addition controls the setting of concrete (Shi, 1996; Odler, 2004).

Therefore management of both fly ash and brines pose a major environmental risk to surface water and land availability if not properly dealt with. However, the pozzolanic properties of fly ash make it suitable for utilisation in agriculture, waste treatment and cement extension (Shehata, 2001; Muriithi et al., 2011; Kruger and Surridge, 2009; Fester et al., 2008; Vadapalli et al., 2008).

Existing literature focuses on mine backfill with thickened tailings (Jewell and Fourie, 2006; Potvin et al., 2005; Benzaazoua et al., 1999) and rarely on water-based fly ash pastes (Steward and Slatter, 2009; Stropnik and Južnič, 1988; Naik et al., 2009). It was therefore scientifically justifiable to investigate the behaviour of

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pastures prepared with brines and fly ash; initial results indicated that contaminants are stabilised in paste (Ilgner, 2006; Mahlaba and Pretorius, 2006; Muntingh et al., 2009; Mahlaba et al., 2008). The current authors investigated the influence of industrial brines on the characteristics of fly ash pastes where it was demonstrated that brine chemistry dominates paste behaviour with a given fly ash type (Mahlaba et al., 2011). These findings suggest that co-disposal of these wastes as a paste backfill material will provide an innovative solution which is environmentally less harmful than their individual disposal.

Paste properties (especially its rheology) are influenced by numerous factors of the materials used (Jewell and Fourie, 2006; Verburg, 2001). The present study sheds light on the influence of fly ash characteristics on the brine-based paste behaviour due to site specificity reported in literature (Jewell and Fourie, 2006; Verburg, 2001). Improved understanding of paste behaviour as a function of both fly ash and brine characteristics will potentially lead to the development of a sound backfill solution as well as geo-technical utilisation opportunities. This manuscript makes a significant contribution towards ensuring the sustainability of the coal processing industry.

2. Materials

2.1. Brines

Brines A and B originate from ion-exchange demineralisation (including regeneration chemicals) and thermal evaporation of water at a South African petrochemical plant, respectively. These industrial brines represent the worst case scenarios of brines from most desalination facilities in terms of chemical composition and salinity, and to a certain degree simulate seawater. The chemical composition of these brines and seawater is shown in Table 1.

2.2. Fly ash

Different fly ashes were collected from two South African power stations combusting different coal types to generate electricity using coal-fired boilers. The elemental and mineralogical composition of these fly ashes, namely, fly ashes A and B is provided in Table 2.

2.3. Examination of physical properties

It is well documented in literature that fly ash characteristics principally depend on coal type and combustion method. Finer ash particles are richer in the glassy phase and are more reactive while coarser fractions are richer in carbon (Ward and French, 2006; Nochaiya et al., 2009; Chancey et al., 2010). Spherical fly ash particles reduce friction between particles and improve workability at lower water demands in a paste (Campbell, 1999; Chindaprasirt et al., 2005). Moreover reduced water demand re-

Table 1
Chemical composition of brines A, B and seawater.

Component	Unit	Brine A	Brine B	Seawater
pH	–	7.4	8.8	8.2–10.0
EC	μS/cm	70,400	124,000	–
Ca ²⁺	mg/l	341	2100	500
Mg ²⁺	mg/l	238	1550	1550
Na ⁺	mg/l	19,227	21,000	12,000
Cl [–]	mg/l	14,668	34,300	22,000
SO ₄ [–]	mg/l	5931	15,200	3000
TDS	mg/l	44,400	108,000	39,806–45,000

#TDS = total dissolved solids.

Table 2
Elemental and mineralogical composition of fly ashes A and B (%).

Component	Fly ash A	Fly ash B
<i>Elemental composition (%)</i>		
SiO ₂	49.7	59.5
Al ₂ O ₃	26.2	28.5
Fe ₂ O ₃	2.7	5.9
CaO	10.5	2.3
MgO	2.1	0.4
SO ₃	0.5	0.4
Na ₂ O	0.7	–
K ₂ O	0.9	1.0
TiO ₂	1.5	1.3
Loss on ignition (LOI)	4.1	–
Other	0.9	0.7
Total	99.8	100.0
<i>Mineralogical phase (%)</i>		
Mullite (Al ₆ Si ₂ O ₁₃)	20.53	28.98
Quartz (α-SiO ₂)	10.24	11.86
Hematite (Fe ₂ O ₃)	0.68	1.43
Lime (CaO)	2.22	0.37
Glassy phase	66.33	57.36
Total	100.0	100.0

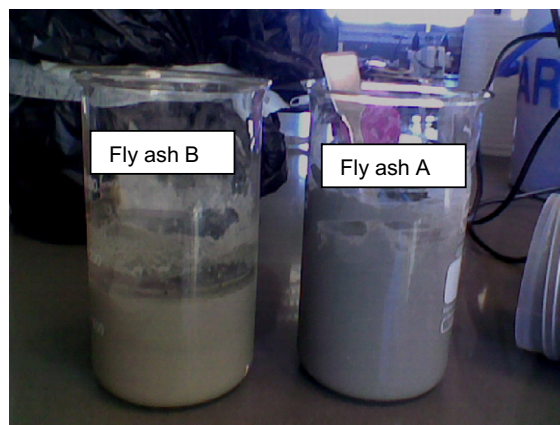


Fig. 1. Picture showing colour difference between fly ash A and B.

sults in a more cohesive paste with minimal bleed formation and low hydraulic conductivity; the cornerstones of good paste (Pagé and Spiratos, 2000; Chindaprasirt et al., 2005; Joshi et al., 1994).

It was considered necessary to discuss physical characteristics of fly ash in addition to chemical properties to enable better interpretation of paste behaviour. The fundamental physical characteristics of fly ash examined are colour, particle size distribution, and particle morphology.

2.3.1. Colour

There is a significant colour difference between the two fly ashes where fly ash A is greyish like ordinary cement and fly ash B is brownish¹ as depicted in Fig. 1. Such a difference can be assigned to an appreciably higher concentration of iron in fly ash B (Table 2) i.e. chemical composition.

2.3.2. Particle morphology

It is illustrated in Fig. 2 that the majority of particles in fly ash A has a spherical morphology while that of fly ash B is rather irregular as depicted in Fig. 3. Therefore pastes prepared with fly ash A

¹ For interpretation of colour in Fig. 1, the reader is referred to the web version of this article.

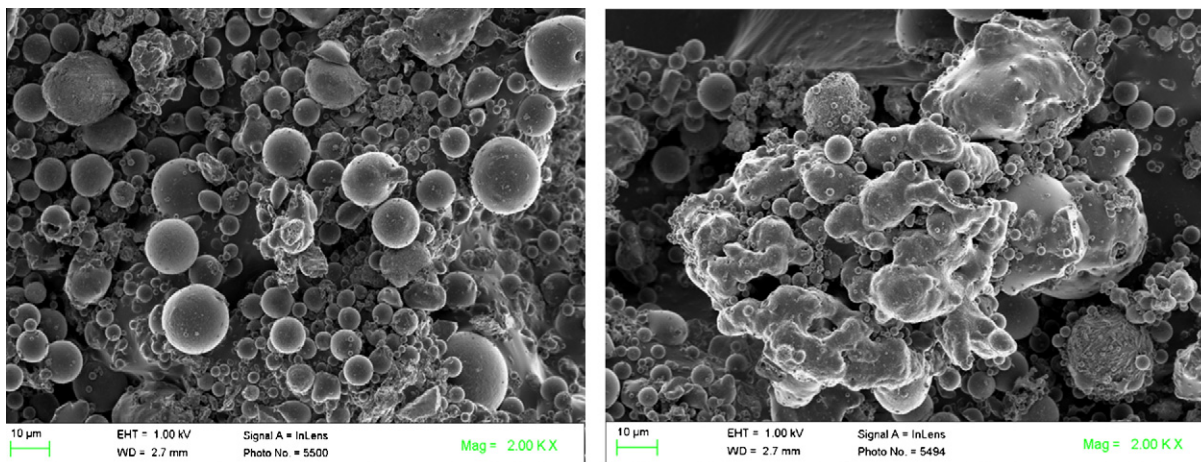


Fig. 2. Typical morphology of fly ash A particles.

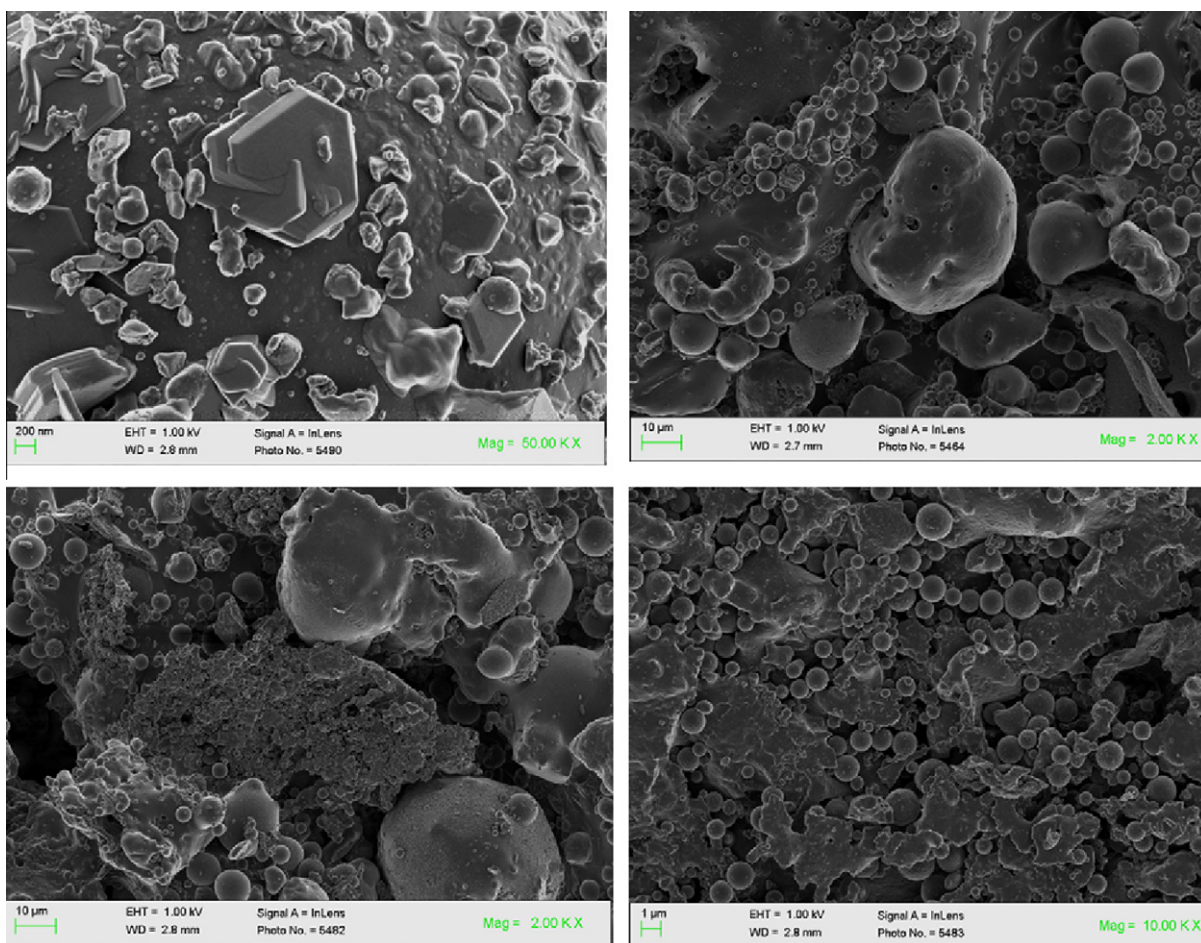


Fig. 3. Characteristic morphology of fly ash B particles.

are expected to be more workable and stronger than those containing fly ash B (Pagé and Spiratos, 2000; Chindaprasirt et al., 2005).

2.3.3. Particle size distribution

Particle size distribution (PSD) profiles of the two fly ashes were determined to indicate basic differences. From Fig. 4 the abundance of particles <20 µm in fly ash can be deduced, which, as a rule of thumb, must exceed 15% for a stable paste formation (Jewell

and Fourie, 2006). It is evident that fly ash A has over 40% while fly ash B has approximately 20% of particles below the threshold particle size, giving the former an advantage to form a better paste.

The PSD data were further classified into different size fractions according to ASTM D 422-63 as shown in Fig. 5. The abundance of clay-sized particles in fly ash A is almost twice that of fly ash B while that of silt-sized particles is 17% higher in the former. As expected the sand-sized particles are significantly higher in fly ash B.

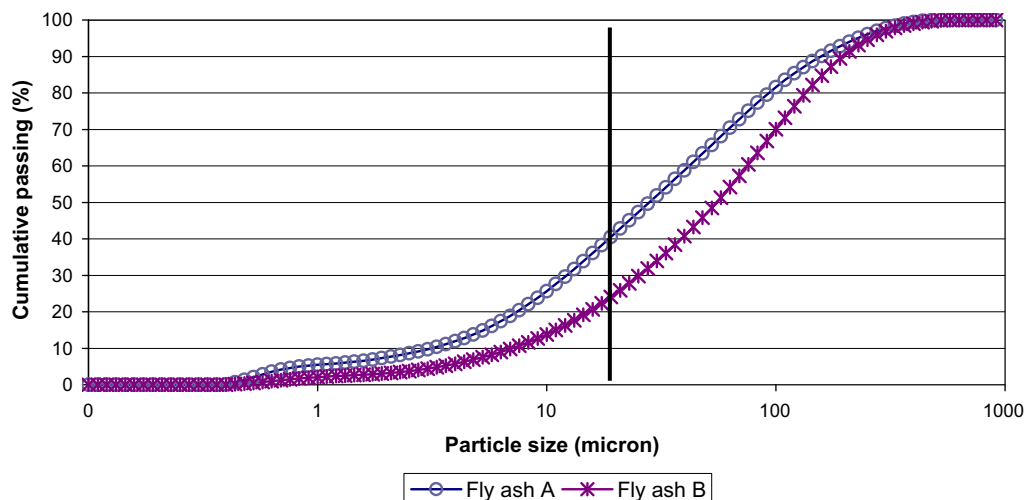


Fig. 4. Particle size distributions of the two fly ashes.

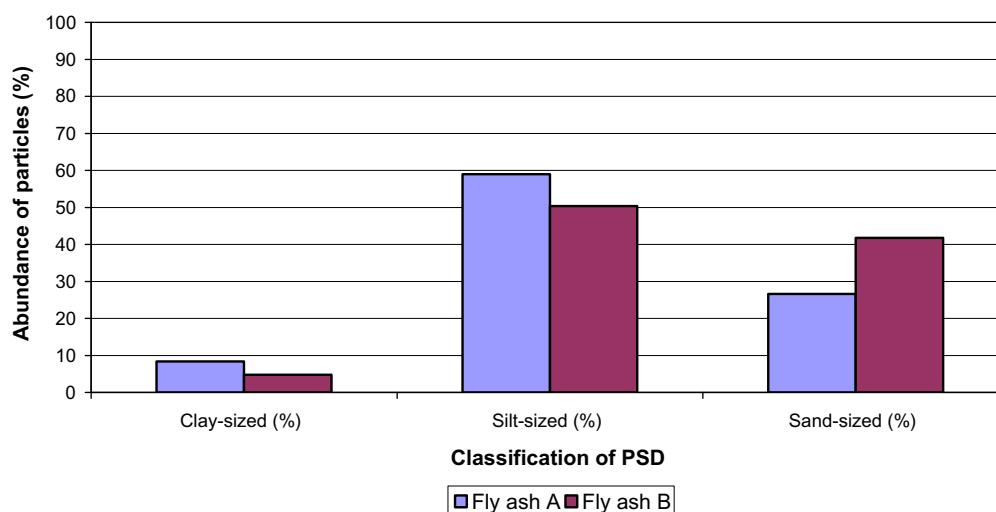


Fig. 5. Particle size distributions of the two fly ashes.

Thus fly ash B is coarser than fly ash A and according to literature finer particles are more reactive than coarser particles which are generally rich in unburned carbon (Chancey et al., 2010; Campbell, 1999; Mahlaba and Pretorius, 2006).

3. Experimental

The role of fly ash as a function of brine variability was studied where workability and compressive strength of paste were used as indicators of the effect. These parameters are essential to determine the suitability of cementitious materials for engineering applications. Naik et al. (2009) emphasise that quick settling of fly ash slurries induces workability challenges substantiated through rheology measurements in the study. Mahlaba and Pretorius (2006) previously reported that brine chemistry can be used to control the hardening of paste during its transportation and save on plasticisers.

3.1. Yield stress

Yield stress of fresh paste was measured after 15 min of maturation using an Anton Paar rheometer set at low vane speeds (Boger et al., 2008) to achieve reproducible data. Material proper-

ties including those discussed in Section 2.3 affect rheological behaviour and maturation time of paste. For instance Naik et al. (2009) used a maturation time of 60 min in fly ash slurries they investigated.

3.2. Unconfined compressive strength

The paste specimens were cured for 28 days before determining the unconfined compressive strength (UCS). A pre-load of 10 N was first applied before data collection commenced at a compression rate of 2.5 mm/min to failure (ASTM C109), using a Z050 Zwick Roell compression machine with 50 kN capacity.

3.3. Effect of fly ash properties

The effect of fly ash properties on the paste behaviour was evaluated by subjecting two types of fly ash to varying brine conditions. Fly ashes were mixed with various amounts of brines A, B, and deionised water to make a series of pastes containing between 62% and 70% fly ash content by mass.

Brine salinity was previously identified as one of the important determinants of paste rheology and strength development (Mahlaba et al., 2011). The response of pastes containing these

fly ashes to increasing salinity will reveal their influence on the resultant paste properties. Brine B was selected to study the effect of salinity on pastes containing the two fly ashes. The dilution of brine B was performed with deionised water to cover a wide range of salinity. The total dissolved solids (TDS) of these solutions were determined at 200 °C. Yield stress and UCS were determined on pastes containing these solutions and 68% fly ash (Mahlaba et al., 2008).

4. Results and discussion

The aim of using fly ash with different chemical properties was to verify whether ash chemistry influences the paste behaviour and test the applicability of results obtained here to similar operations. This section presents the results obtained for pastes containing the two fly ashes as a function of brine variance. The emphasis was placed on both the rheological and mechanical characteristics of the resultant pastes.

4.1. Rheological behaviour

The rheological results presented in Fig. 6 demonstrate that the workability of pastes prepared with fly ash B is not influenced by variability in brine composition. All pastes with fly ash B gave a

yield stress of approximately 40 Pa. On the contrary, pastes derived from fly ash A with brine A and deionised water gave a yield stress of approximately 80 Pa which is twice that of pastes bearing fly ash B. A substantially higher yield stress of 210 Pa was achieved by paste containing fly ash A and brine B. Mahlaba et al. (2011) discovered that a salinity of brine must not exceed 60,000 mg/l for pastes prepared with fly ash A to remain pumpable with centrifugal pumps.

Fig. 7 also shows a negligible response of fly ash B bearing pastes to changes in brine salinity. In contrast, fly ash A pastes demonstrated an exponential increase in the yield stress when TDS in brine exceeded 40,000 mg/l. The increase in the yield stress as a function of brine salinity for pastes derived from fly ash A indicates the occurrence of early hardening reactions (Campbell, 1999). This behaviour was not exhibited by pastes bearing fly ash B which is indicative of poor reactivity and thus less strength development is anticipated for pastes containing fly ash B.

4.2. Unconfined compressive strength

The UCS results of pastes containing different fly ash types exposed to varying brines are depicted in Fig. 8. It is illustrated that fly ash A responded to different brines whereas fly ash B was less reactive within 28 days. The UCS of fly ash B pastes was below

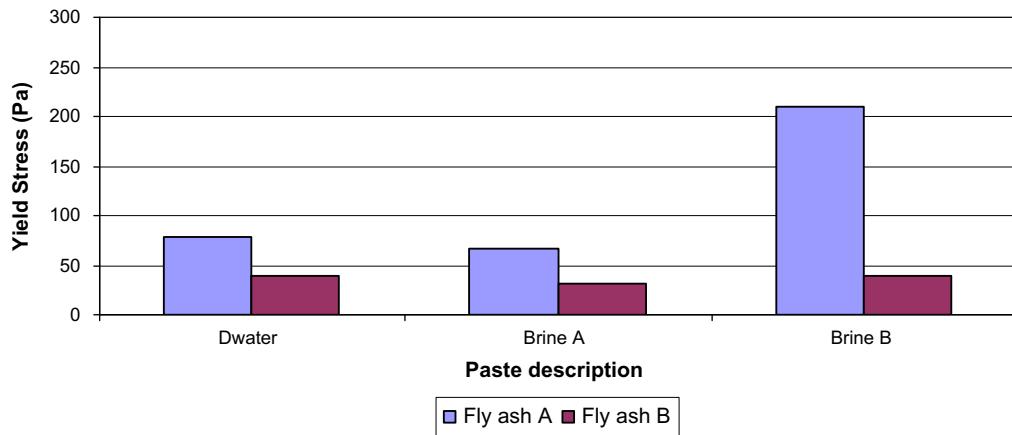


Fig. 6. Effect of fly ash properties on workability.

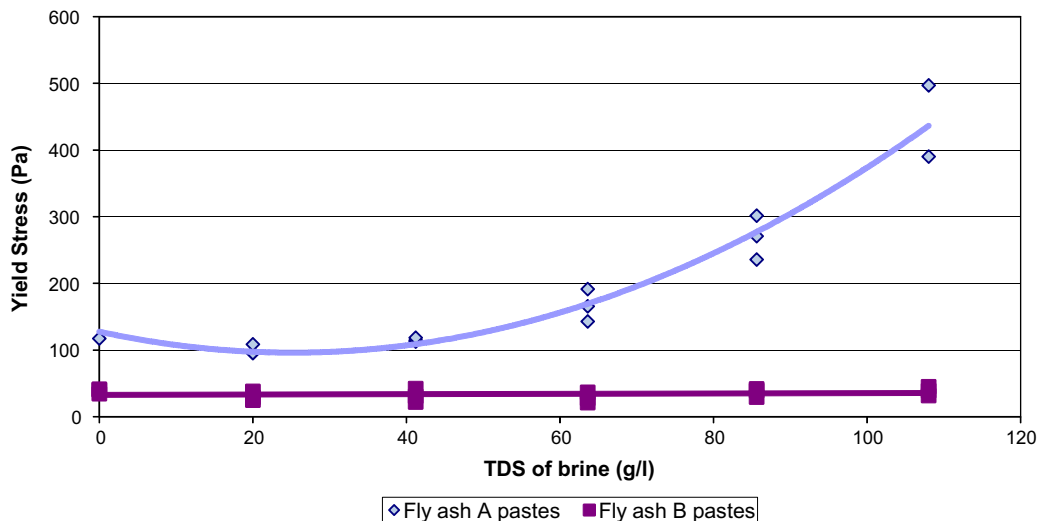


Fig. 7. Rheological response of pastes to salinity per fly ash type.

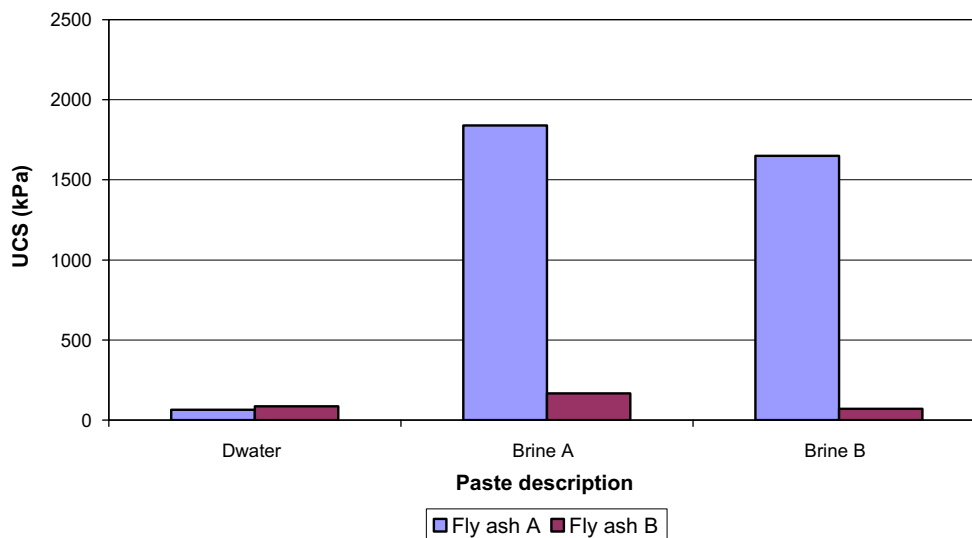


Fig. 8. Influence of fly ash on the strength development of pastes prepared with varied brines.

200 kPa whereas pastes containing fly ash A reached 1840 kPa and 1650 kPa with brine A and brine B, respectively. The results obtained with fly ash A bearing pastes suggest that brine chemistry affects the mechanical properties of paste, not necessarily according to salinity (Mahlaba et al., 2011). Pastes prepared with brines and fly ash A significantly exceeded a minimum UCS of 200 kPa suggested by Potvin et al. (2005) for mine backfill. Mahlaba (2007) showed that the pH of paste remained high even when acidic brines were used due to high alkalinity of fly ash which agrees with its use of neutralising acid mine drainage (Vadapalli et al., 2008; Gitari et al., 2006; Shang and Wang, 2005; Benzaazoua et al., 2002; Fester et al., 2008).

The distinction exhibited by two fly ashes was attributed to the mineralogical differences especially lime deficiency and lower content of the amorphous phase in fly ash B (Ward and French, 2006; Donahoe, 2004). Furthermore, higher degree of fineness and sphericity in fly ash A contributes to its superior performance on desired paste characteristics. Nevertheless, the use of additives such as lime and cement can ameliorate the reactivity of fly ash B for paste backfill systems (Giergiczny, 2004; Shi, 1996).

This study has clearly shown the benefits of utilising brine instead of water in fly ash paste backfill applications. The ultimate goal and benefit to stakeholders is ensuring water conservation and sustainable energy generation from coal.

5. Conclusions and recommendations

The important findings of this study reveal that:

- Paste backfill of brines with fly ash is possible if suitable materials are used.
- The paste properties are site-specific and generalisation may be too risky and inaccurate.
- Paste behaviour is better explained by considering chemical, mineralogical and physical properties of the fly ash in addition to brine chemistry of the mixing water.
- Fly ash characteristics seem to play a more important role than brine chemistry because fly ash is the major component and provides stabilisation of contaminants in the paste disposal application.

This manuscript was extracted from the PhD thesis of the lead author. Future research will have to investigate ways of ameliorat-

ing the characteristics of fly ash B to achieve similar paste properties as fly ash A. The leachability of resultant pastes will be a critical factor in determining the environmental impact of this promising application.

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