ARTICLE IN PRESS

Minerals Engineering xxx (2011) xxx–xxx

Contents lists available at ScienceDirect

Minerals Engineering

journal homepage: www.elsevier.com/locate/mineng

Evaluation of workability and strength development of fly ash pastes prepared with industrial brines rich in $SO_4^{=}$ and Cl^{-} to expand brine utilisation

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ARTICLE INFO

Article history:
Received 26 March 2011
Accepted 19 May 2011
Available online xxxx

Keywords:
Brine
Water salinity
Coal fly ash
Paste backfill
Waste management

ABSTRACT

Anthropogenic pollution is an unavoidable consequence of both producing energy from coal and desalination of water. Coal ash and brines are partially utilised due to vast volumes and stringent legal environmental requirements. Therefore innovative management for these wastes is essential. This manuscript presents the initial results of research showing that brine chemistry dominates the behaviour of fly ash pastes. The outcome could expand the utilisation of brines in mortars and mass concrete to conserve potable water. The tests involved varying paste consistency and brine characteristics. The results demonstrated that chemical composition of brine plays a more important role than salinity in determining both paste rheology and strength development. An optimum brine salinity range for pastes was obtained with a specific fly ash. The results suggest that an opportunity exists for utilising industrial brines rich in Cl^{-} and $SO_4^{=}$ as mixing waters in the co-disposal or mine backfilling with fly ash pastes. This would reduce operational costs and liability of energy generation from coal.

1. Introduction

The scarcity of potable water and escalating energy demands are arguably the major contributors to anthropogenic pollution in terms of saline brines and coal fly ashes, respectively. The management of industrial brines resulting from water recovery poses an environmental concern especially inland (Nassar et al., 2008; Souilah et al., 2004; Vedavyasan, 2001); where the option of oceanic disposal is often uneconomical or may have detrimental effects on marine life over the long-term (Souilah et al., 2004; Ahmed et al., 2003; Korngold et al., 2009).

Conventional disposal of brines along with fly ash as a slurry uses copious volumes of water which makes the transportation simple. The excess water required presents a risk of leaching heavy metals and contaminate fresh water resources and soil. Paste disposal increases the ratio of fly ash to water which will conserve water and reduce leaching. The co-disposal of brines with fly ash has been proposed by several researchers as a means to mitigate the environmental footprint (Palarski et al., 2011; Mahlaba et al., 2008; Muntingh et al., 2009; Ilgner, 2002; Mahlaba, 2007). Transportation of fly ash paste to a disposal site is a major challenge to operators and technology suppliers (Steward and Slatter, 2009; Naik et al., 2009; Jewell and Fourie, 2006). This is often due to the sensitivity of rheology to fly ash properties such as particle size distribution, particle morphology, and mineralogy (Boger et al., 2008). The matter is further exacerbated in cases where brines exhibit variable characteristics.

The variability of brines as well as failure to meet legal environmental requirements restrict their potential utilisation in applications such as a carrier medium of solid wastes, crystallisation of marketable salts, and mixing water in concrete. Literature focuses on the deleterious 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 (Medveščček et al., 2006; Collepardi, 2003; Klemm, 1998). These anions pose a durability issue if they come from the external environment such as the interaction of concrete with seawater.

Nevertheless, a few researchers have investigated the utilisation of seawater as mixing water in concrete and discovered that resultant concrete was stronger than a control which was prepared with potable water (Borsoi et al., 2009; Akinkurolere et al., 2007). The major components in seawater are chloride, sodium, calcium and sulphate (Taylor and Kuwairi, 1978). The examination of most saline brines also demonstrates that calcium, sodium, sulphates and chlorides are the major constituents (Ahmed et al., 2003; Ravizky and Nadav, 2007; Mooketsi et al., 2007; Koch, 2002). Therefore close similarity exists between saline brines and seawater. Furthermore, the use of Cl-bearing compounds to accelerate strength development and improve mechanical properties is common

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doi:10.1016/j.mineng.2011.05.015

practice in concrete (Akinkurolere et al., 2007; Taylor and Kuwairi, 1978; Shi, 1996). Preliminary work by Mahlaba (2007) and Mahlaba and Pretorius (2006) indicated that, compared to water, brines have an advantageous effect on the workability of fly ash pastes.

The shortage of potable water and similarity between industrial brines and seawater motivated this research which can hopefully contribute towards sustainability. The primary objective was to investigate the role of brine chemistry on the behaviour of fly ash pastes in order to develop a sound co-disposal method for these wastes. Achieved results could also moot the extension of brine utilisation as mixing water in cementitious materials e.g. concrete and mortars.

State-of-the-art techniques were employed to investigate the effect of brine characteristics on the paste behaviour. This manuscript sets the scene and presents the preliminary results on how brine chemistry influences the workability and strength development of fly ash pastes.

2. Materials

2.1. Brines

Brine A and brine 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 typical seawater are shown in Table 1. These industrial brines were used to study the influence of brines on the rheological and hardening properties of paste. Deionised water was used as a control and also to present the best case scenario of the mixing water for paste preparation without chemical influence. Salinity is expressed in terms of total dissolved solids (TDS) in this manuscript.

2.2. Fly ash

Fly ash was obtained from a South African petrochemical plant which combusts low-grade bituminous coal for the production of steam and electricity to meet its process requirements. The elemental composition of this fly ash is provided in Table 2.

The mineralogical data of the fly ash is presented in Table 3. The presence of lime and high content of glassy phase indicate that this fly ash should be reactive upon contact with water to form materials with cementitious properties (Ward and French, 2006; Kolay and Singh, 2001; Donahoe, 2004). This is referred to as a pozzolanic reaction.

Table 1
Chemical composition of brine A, brine B and seawater (Mahlaba et al., 2011a).

Component	Unit	Brine A	Brine B	Seawater
pH	–	7.4	8.8	8.2
EC	μS/cm	70,400	124,000	–
Ca	mg/L	341	2100	500
Mg	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

Table 2
Elemental composition of fly ash (%).

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	TiO_2	SO_3	LOI[a]	Other	Sum
49.7	26.2	2.7	10.5	2.1	0.9	0.7	1.5	0.5	4.1	0.9	99.8

[a] LOI = loss on ignition.

Table 3
The mineralogy of fly ash used in the study (%).

Phase	Chemical formula	Abundance (%)
Mullite	$Al_6Si_2O_{13}$	20.53
Quartz	α-SiO_2	10.24
Hematite	Fe_2O_3	0.68
Lime	CaO	2.22
Glassy phase	N/A	66.33
Total	N/A	100.00

Table 4
The specimen mixes used in the study (% m/m).

Fly ash content (%)	Brine type (%)
62	38
64	36
66	34
68	32
70	30

3. Experimental

Workability and compressive strength are the critical parameters which determine the suitability of cementitious materials for engineering applications. Rheology measurements are commonly used in a variety of applications including construction, waste management and food industry to study the flow behaviour of viscous materials such as pastes (Jewell and Fourie, 2006; Kwak et al., 2005; Huynh et al., 2006; Nguyen and Boger, 1998; Nguyen et al., 2006). Yield stress was used as a measure of the workability of pastes. Yield stress defines the minimum shear stress required to initiate significant flow. A value of 200 Pa was selected as the maximum threshold value of yield stress for paste. This figure coincides with the limit for centrifugal pumps (Boger et al., 2006) falling within a yield stress range of 100–500 Pa suggested by Hallbom (2010) for paste backfill materials.

An unconfined compressive strength of 500 kPa is used in this work as the lower limit of strength applicable to paste. This is higher than the minimum values recommended by other researchers (Potvin et al., 2005; Bouzalakos et al., 2008).

3.1. Yield stress

An Anton Paar rheometer was used to determine yield stress. To improve the reproducibility measurements were taken after 15 min of paste wetting at a low vane speed of 0.5 rpm (Boger et al., 2008).

3.2. Unconfined compressive strength

The paste specimens were wrapped in plastic and allowed to cure 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).

3.3. Effect of brine chemistry

Fly ash was mixed with various amounts of brine A, brine B, and deionised water to make a series of pastes containing between 62% and 70% fly ash content on a mass basis. Details on the selected range are given by Mahlaba (2007). Table 4 gives the specimen mixes investigated in the study.

3.3.1. Effect of salinity

Brine B was selected to study the effect of salinity on paste properties by virtue of its composition and high salinity (Table 1). The dilution of brine B was performed with deionised water as follows; 20:80 (20%), 40:60 (40%), etc. with 100% being brine B in its original form. The TDS of these solutions was determined at 200 °C. Yield stress and UCS were determined on pastes containing these solutions and 68% fly ash, which is a suitable paste consistency for evaluating changes in the paste behaviour (Mahlaba et al., 2008; Mahlaba and Pretorius, 2006).

4. Results and discussion

The presentation of rheological data in this manuscript necessitated the adoption of the following equation:

$$TS = TSS + TDS \quad (1)$$

where TS = total solids (mg/L), TSS = total suspended solids i.e. fly ash content (mg/L), TDS = total dissolved solids in the brines (mg/L).

Deionised water is abbreviated as Dwater in the graphs.

4.1. Effect of brine chemistry

The results illustrated in Fig. 1 demonstrate that total dissolved solids (TDS) of a brine solution affects the yield stress of paste. The higher the salinity of brine the less workable the paste becomes for a given fly ash content and vice versa. Yield stress for pastes containing brine B was approximately 200 Pa at 68% fly ash content while for brine A and deionised water the yield stress of the pastes was below 100 Pa. The yield stress curves of brine A and deionised water pastes diverge after 70% fly ash content probably because of solid saturation. Conversely, brine B pastes diverged at as low as 64% fly ash content probably because it has a higher TDS.

The curves are almost superimposed when TDS is taken into account in the calculation of total solids (TS) in pastes as shown in Fig. 2. This observation demonstrates that salinity influences the workability of pastes prepared with different brines. It is therefore concluded that TS provides better basis for evaluating the workability of pastes than TSS according to Fig. 1. It is also possible to predict the yield stress of a paste in terms of TS by fitting an equation in Fig. 2.

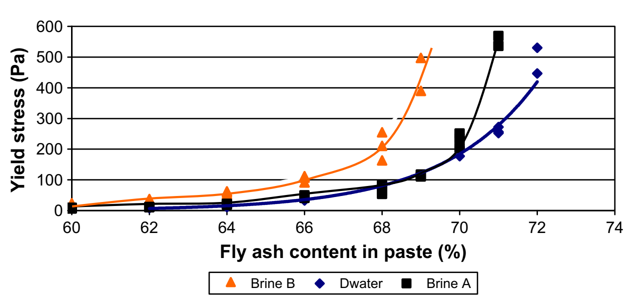

Fig. 1. Yield stress for various pastes as a function of fly ash (TSS).

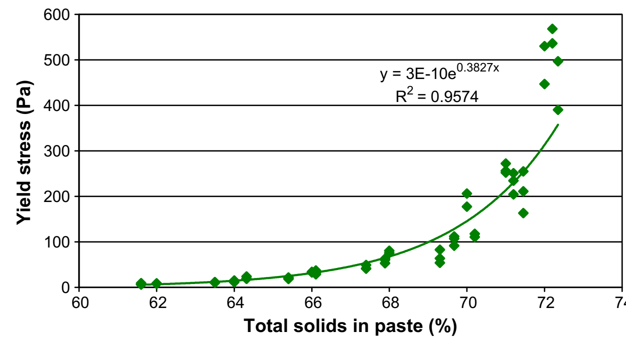

Fig. 2. Yield stress for various pastes as a function of total solids (TS).

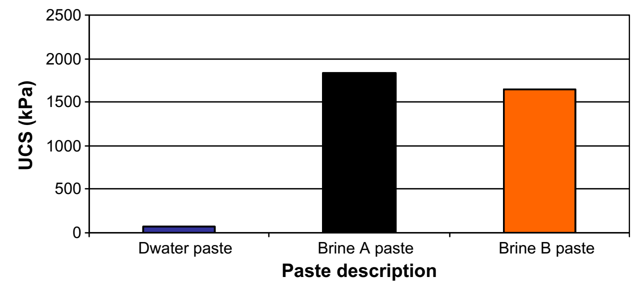

Fig. 3. Influence of brine type on unconfined compressive strength.

The results of UCS for pastes containing different brines are presented in Fig. 3. Brine A gave the highest UCS value of 1840 kPa followed by brine B with 1650 kPa whereas deionised water gave the lowest UCS value of 65 kPa. Other researchers have also made a similar observation and concluded that operational costs of coal mining can be reduced by using saline mine waters (Palarski et al., 2011). These results indicate that brine chemistry affects the mechanical properties of paste, not necessarily according to salinity. This highlights the importance of chemical composition of the brine in the paste properties as reported by Mahlaba (2007) and Mahlaba and Pretorius (2006).

4.2. Influence of brine salinity

A need was identified to decouple the effect of chemical composition from that of the brine salinity since both parameters influenced the results. The results of yield stress as a function of brine salinity (as TDS) used during paste preparation are presented in Fig. 4 demonstrating that the salinity of brine influences the workability of paste.

It is therefore concluded that the use of a more saline brine will result in a less workable paste in a given fly ash-to-brine ratio. This may elevate the energy requirements for transportation and susceptibility of paste to pipeline blockages. For instance, a paste

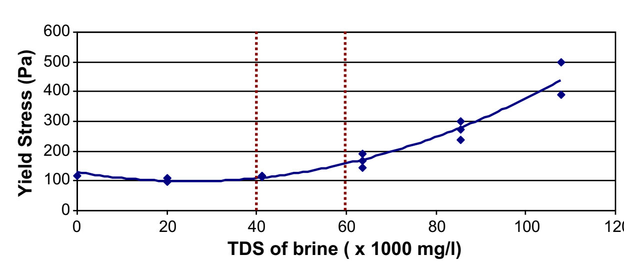

Fig. 4. Effect of salinity on yield stress (68% fly ash).

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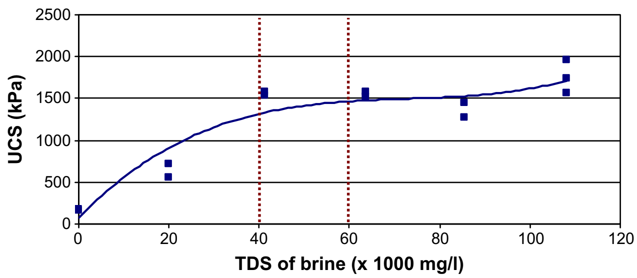

Fig. 5. Effect of salinity on compressive strength.

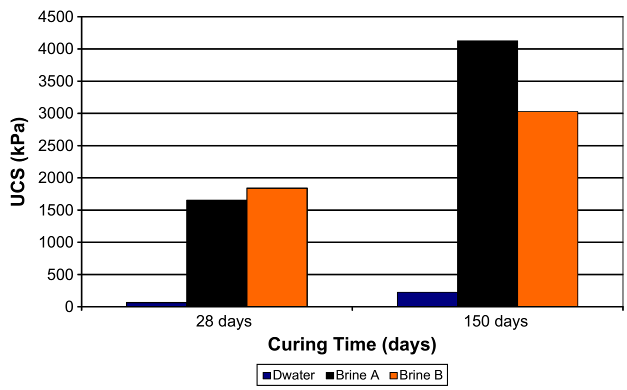

Fig. 6. Effect of curing time on UCS development of brine containing fly ash pastes.

containing 68% fly ash and brine with ⩽40,000 mg/L TDS has a yield stress of 120 Pa whereas a paste containing the equivalent fly ash content and 85,000 mg/L saline brine gives a yield stress of 280 Pa. Although brine salinity has a significant influence on the workability of paste, the salinity range of 40,000–60,000 mg/L provides pastes with comparable workability.

The results of UCS for pastes prepared with a fixed fly ash content and brines of varying salinity are presented in Fig. 5. It is apparent that there is an increase in UCS with salinity up to 40,000 mg/L TDS beyond which no further strength gain was achieved. These results suggest that a salinity of at least 40,000 mg/L in a specific brine composition is required for optimum strength gain.

The consideration of both yield stress and UCS data demonstrates that salinity has more influence on workability than on the compressive strength achieved. The use of brine with 40,000 mg/L TDS gave the same mechanical properties as brine containing 85,000 mg/L TDS while yield stress increased rapidly from 120 to 280 Pa. Therefore the operating envelope in terms of salinity lies between 40,000 and 60,000 mg/L TDS as marked in Figs. 4 and 5.

Some authors report that the use of saline waters in paste preparation gives higher initial UCS compared to potable water which then decreases after 28 days of curing (Saw and Villaescusa, 2011; Kermani et al., 2011). However, the present authors found contradictory results where UCS of pastes prepared with brines continued to increase within 150 days (Fig. 6). The distinction between the present study and the findings made by Saw and Villaescusa (2011) and Kermani et al. (2011) can be ascribed to site-specific characteristics of paste (Jewell and Fourie, 2006; Mahlaba et al., 2011a).

The UCS values of pastes containing brine B and brine A increased from 1650 and 1840 kPa to 3000 and 4100 kPa, respectively as illustrated in Fig. 6. The increase in UCS within 150 days suggests a significant extent of reactions taking place between fly ash and brines e.g. formation of C–S–H and hydrated gehlenite (Mahlaba et al., 2011b).

Compared to brine pastes, no appreciable improvement in the UCS of pastes bearing deionised water was achieved since it only rose from 65 to 225 kPa. This difference in behaviour indicates that ionic components present in the brine have a profound influence in the strength development of fly ash pastes (Akinkurolere et al., 2007; Taylor and Kuwairi, 1978).

The article has confirmed that the utilisation of brines has benefits on the workability and strength development of pastes. Due to the complexity of these brines it is, however, not clear which constituents present in industrial brines and seawater are responsible for the observed paste properties. An in-depth investigation of prevalent brine components on the paste behaviour should be conducted.

5. Conclusions

The findings of this research can be summarised as follows:

(a) Total solids of paste (rather than fly ash content) provide better means of assessing and predicting workability.
(b) An optimum range of salinity for brine B was found between 40,000 and 60,000 mg/L for acceptable workability and adequate strength development when mixed with fly ash. It must be noted that this range may change if a different ash or brine is used.
(c) The increase in UCS was observed within 150 days of paste curing.
(d) It is concluded that industrial brines rich in chlorides and sulphates can be beneficially used in cementitious products such as mortars and mass concrete. This discovery substantiates the use of seawater in concrete as observed by other researchers.

This work has demonstrated that brines can be co-disposed with fly ash as paste backfill material (based on physical properties) which has countless benefits to sustainability. Moreover, brines can potentially be utilised as mixing waters in concrete without steel reinforcement. The significance of brine utilisation is that it will minimise the consumption of the already scarce water resources as well as disposal costs.

Acknowledgements

The authors would like to thank SASOL Technology R&D (Pty) Ltd. for sponsoring this project and granting permission to publish this work. The University of Pretoria and Richonne Consulting are highly acknowledged for the support and healthy debates which led to the success of the project. This manuscript emanated from the PhD thesis of the lead author.

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