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Comparative Assessment of the Leaching Properties of Fly Ash and Phosphogypsum Disposed of in South Africa

Jabulani Matsimbe^{1, 2, 3*}, Megersa Dinka¹, David Olukanni⁴, Innocent Musonda²

¹Department of Civil Engineering Science, Faculty of Engineering and the Built Environment, University of Johannesburg, Johannesburg 2006, South Africa

²Centre for Applied Research and Innovation in the Built Environment (CARINBE), Faculty of Engineering and the Built Environment, University of Johannesburg, Johannesburg 2092, South Africa

³Department of Mining Engineering, Malawi University of Business and Applied Sciences, P/Bag 303, Chichiri, Blantyre3, Malawi

⁴Department of Civil Engineering, Covenant University, 10 Idiroko Road, Ota, Ogun State, Nigeria
jmatsimbe@poly.ac.mw; mdinka@uj.ac.za; david.olukanni@covenantuniversity.edu.ng; imusonda@uj.ac.za

Abstract - Fly ash (FA) and phosphogypsum (PG) are readily available industrial waste materials commonly disposed of in landfills. Knowledge of the leaching behavior of FA and PG can help us understand how to diminish their adverse effects on the ecosystem. However, there is limited research on the leaching properties of the FA and PG disposed of in South Africa. This study aims to comparatively assess the leaching properties of FA and PG disposed of at Eskom Duvha Power Station in Mpumalanga and Foskor Ltd in Phalaborwa, South Africa. The toxicity characteristic leaching procedure (TCLP) was used to obtain the leachates which were further analyzed by the inductively coupled plasma-mass spectrometer (ICP-MS). An XRF spectrometer was used to find the chemical composition of FA and PG. It was found that both FA and PG are non-hazardous since their leachate concentrations are within the US EPA TC, US EPA land disposal regulation, and EU waste acceptance at landfills. The solubility and mobility of As and Se depend on the alkaline concentration of the solution. The As adsorption rate escalates in Fe oxides and its mobility depends on the waste chemistry and pH. The chemical elements present in FA and PG i.e., Al³⁺, Si⁴⁺, Fe²⁺, Ca²⁺, instead of being leached into the ground can be incorporated into cement for construction applications. The present study output contributes knowledge on leaching properties of FA and PG in South Africa and can help policymakers create informed solid waste management frameworks and understand the potential of reusing in construction and recovering the elements of economic interest (e.g., Zn, Cu, Fe, P). The recycling of FA and PG for use as construction materials can help address landfill leaching problems and promote circularity.

Keywords: Circularity, heavy metals, leachate immobilization, recycling, supplementary cementitious materials, sustainability

1. Introduction

Fly ash and phosphogypsum are waste materials generated by coal-fired thermal power stations and phosphoric acid plants [1], [2]. FA is grouped into Class F or Class C depending on the oxide composition which comprises CaO, Al₂O₃, SiO₂, and Fe₂O₃ [3]. PG is composed of gypsum (CaSO₄·2H₂O) and is a by-product of the wet acid process – a reaction between fluorapatite (Ca₅(PO₄)₃F) and sulphuric acid (H₂SO₄) that produces phosphoric acid (H₃PO₄) [4]. Globally, the annual production of FA is around 1 billion tonnes and that of PG is around 300 million tonnes but only about 15 – 50% is reused by industries [5]-[7]. The remaining 50 – 85% of FA and PG is usually disposed of in landfills leading to environmental pollution and incurring treatment/disposal costs. South Africa alone contributes 40 million tonnes of FA and 35 million tonnes of PG per annum [8]. The recycling of FA and PG can help reduce landfill exposure to weathering processes, eliminate waste disposal costs, and provide alternative construction materials for sustainable infrastructural development in line with sustainable development goals (SDGs) [9], [10] and the circular economy [11], [12]. However, to ensure that the FA and PG are fully reutilized in infrastructural development, it is imperative to examine their leaching properties. The landfilled waste is exposed to water which seeps through it producing a leachate. Several researchers have studied the leaching mechanisms of FA and PG worldwide. For instance, Urgulu [13] studied

the leaching characteristics of FA at a Power Station in Turkey. The results showed that the leachate amounts of Fe, Mn, S, K, Na, Pb, and Ca were higher than Cr, Co, Cu, Mg, Cd, and Zn. In another study, Gitari et al. [14] investigated the leaching properties of SASOL Sekunda fly ash (SFA) and Eskom Tutuka fly ash (TFA). They found that the leachate concentration of Cr, Se, Pb, Cd, and As, in the FA satisfied the South African Target Water Quality Range (TWQR). The concentration of Zn, Cr, Pb, Y, Cu, Ni, Rb, and Co in TFA was higher than that of SFA. The amounts of La, Zr, Ba, Ce, Sr, and Nb were higher in SFA than in TFA. The difference in leachate amounts of the two FA was attributed to the mineralogy and elemental composition of the feed coal. The study further revealed that the solution pH significantly affected the leaching of elements from FA, which agrees with [13]. Zhang et al. [15] utilized FA as an additive to co-treat oily sludge and evaluated the leaching behaviour after solidification. The results showed that the FA controlled the melting behaviours of oily sludge and supported the heavy metals solidification providing a sustainable disposal approach for oily sludge treatment. Similarly, Dacuba et al. [16] found that adding FA in clay-fired bricks decreased the movability of Mo and immobilized Se and Cr. Bouargane et al. [17] studied the recovery of Na₂SO₄, CaCO₃, and Ca(OH)₂ from PG in Morocco. The compounds were processed through the exact stoichiometric reactant proportions and characterized by XRD. The results showed that the dissolution of 5g of PG in 0.3M of NaOH led to the precipitation of Ca(OH)₂ and Na₂SO₃ as a function of time, pH, and electrical conductivity. The addition of CO₂ to an aqueous slurry of portlandite, Ca(OH)_{2(aq)}, at a flow rate of 10 mL/min for 3 h led to the precipitation of CaCO₃. The evaluation of heavy metals/trace elements showed that Portlandite (Ca(OH)₂) and Calcite (CaCO₃) had somewhat similar concentrations to the raw PG except for Thenardite (Na₂SO₄) which had negligible concentrations.

FA and PG are readily available industrial waste materials that are commonly disposed of in landfills. Prolonged landfilling or stockpiling of FA and PG can potentially release dust and some amounts of leachate into the environment. The recycling of FA and PG for use as construction materials can help address landfill leaching problems and promote circularity. Knowledge of the leaching behavior of FA and PG can help us understand how these industrial waste materials can behave if used for construction purposes whilst diminishing the adverse effects on the ecosystem. However, there is limited research on the leaching properties of the FA and PG disposed of in landfills in South Africa. This study aims to comparatively assess the leaching properties of FA and PG disposed of in landfills at Eskom Duvha Power Station in Mpumalanga and Foskor Ltd in Phalaborwa, South Africa. The research question addressed is how the FA and PG leaching properties compare with regulatory limits to assess compliance and justify their applicability for sustainable construction purposes. Proper disposal and management of FA and PG can help address environmental pollution from landfill leachates and provide alternative construction materials for infrastructural development.

2. Materials and Methods

FA and PG collected from Eskom Duvha Power Station in Mpumalanga and Foskor (pty) Ltd in Phalaborwa, South Africa, were used. Duvha Power Station, with coordinates 25°57'40''S / 29°20'19''E, is a coal-fired thermal power plant run by Eskom. Foskor (pty) Ltd, with coordinates 23°57'06''S / 31°08'15''E, is a vertically integrated producer of phosphate-based granular fertilizer and phosphoric and sulphuric acid. Representative samples were obtained from the bulk sample using a riffle splitter (Labotec Model: 600) for testing as per ASTM D6913 [18] after which it was dried for 24 hrs in the oven. A sample size (n) of 3 duplicates for FA and PG was used per test (chemical composition, pH, and leaching) after which a mean and standard deviation were calculated. Statistical analysis was done in MS Excel to determine the mean (\bar{x}) and sample standard deviation (s) for n number of observations (x_i) in chemical composition, pH, and leachates:

$$\bar{x} = \frac{\sum_{i=1}^n(x_i)}{n} \quad (1)$$

$$s = \sqrt{\frac{\sum_{i=1}^n(x_i - \bar{x})^2}{(n-1)}} \quad (2)$$

An XRF spectrometer (Rigaku ZSX PRIMUS II) was used to find the chemical composition of FA and PG based on these elements: Fe₂O₃, Al₂O₃, SiO₂, MgO, CaO, MnO, P₂O₅, K₂O, Na₂O, SO₃, and TiO₂, as well as LOI. The Class of FA depends on the CaO content and the sum of SiO₂ + Al₂O₃ + Fe₂O₃ following ASTM C618 [3]. The pH was

determined using a pH meter (HANNA HI98130 Combo) per ASTM D4972 [19]. Distilled water prepared using a water distiller (Labcon WS01-04) was used in the preparation of all test specimens following ASTM D1193 [20].

Batch leaching test based on the toxicity characteristic leaching procedure (TCLP) [21], [22] was done to determine the leachate concentrations and thus examine whether the FA and PG are hazardous/non-hazardous depending on their toxicity characteristic (TC). The FA, PG, and FA+PG mixture was milled and leached with glacial acetic acid/sodium hydroxide solution (pH 4.93 ±0.05) at a solid/liquid ratio of 1:20. The samples were stirred for 18 hours at ambient temperature and filtered to obtain the extract. Different proportions of FA:PG were prepared at increments of 10wt% PG to the mass of FA. The optimal FA-PG mix was obtained at 70 wt% FA: 30 wt% PG prepared at 37% optimum moisture content (OMC) and then heat cured at 40°C for 7 days. The concentration of heavy metals and/or trace elements i.e., Mercury (Hg), Arsenic (As), Nickel (Ni), Zinc (Zn), Cadmium (Cd), Lead (Pb), Copper (Cu), Chromium (Cr), Cobalt (Co), Iron (Fe), Selenium (Se), Manganese (Mn), and Phosphorus (P), were calculated by the inductively coupled plasma-mass spectrometer (PerkinElmer NexION ICP-MS). To assess compliance, the measured leachate concentrations in FA, PG, and FA+PG composite were compared to the US EPA TC limits [23], US EPA land disposal regulation (LDR) limits [24], EU waste acceptance at landfills [25], and the South African DWAF target water quality (TWR) [26]. Due to its greater speed, precision, and sensitivity, the ICP-MS is commonly used to detect heavy metal and/or trace element concentrations [27]. The sample is ionized by the inductively coupled plasma and exposed to a mass spectrometer for separation and detection of different metal and/or nonmetal ions [28], [29]. A similar technique of TCLP and ICP-MS was used by Authors [30]-[32].

3. Results and Discussion

3.1. Chemical composition

Table I shows the chemical composition of FA and PG. A sample size (n) of 3 each for FA and PG was tested and the mean ± standard deviation was calculated. FA is laden with SiO₂ (51.98%), Al₂O₃ (22.80%), Fe₂O₃ (10.02%), and CaO (6.41%) while PG is laden with SO₃ (44.08 %) and CaO (29.17%). The loss of ignition (LOI) equal to 3.83 does not exceed the LOI limit of 6% specified in ASTM C618 [3] implying fly ash with low unburnt carbon which can be attributed to good boiler performance during coal burning. A higher LOI reduces the fly ash quality and limits its applicability in cement/concrete since it affects durability [33]. Furthermore, the fly ash moisture content (MC) of 0.4% and SO₃ content of 0.24 wt% are within the acceptable ASTM C618 requirements of MC < 3% and SO₃ < 5%.

Table I. Chemical composition (wt. %) of FA and PG (mean ± SD)

Compound	FA	PG	Compound	FA	PG
SiO ₂	51.98±0.37	0.65±0.01	TiO ₂	1.48±0	
Al ₂ O ₃	22.80±0.15	0.08±0	MnO	0.07±0.01	0.03±0
Fe ₂ O ₃	10.02±0.43	0.07±0.01	BaO	0.13±0	
CaO	6.41±0.06	29.17±0.18	NiO	-	
MgO	1.39±0.04	0.15±0	V ₂ O ₅	-	
Na ₂ O	-		Cr ₂ O ₃	-	
K ₂ O	0.53±0.02		F	-	2.15±0.08
SO ₃	0.24±0	44.08±1.21	LOI	3.83±0.6	21.90±0.12
P ₂ O ₅	0.76±0.2	1.271±0.4	MC	0.4±0.02	8.93±0.09

ASTM C618 classifies FA into either Class F or Class C depending on the content of $\text{SiO}_2+\text{Al}_2\text{O}_3+\text{Fe}_2\text{O}_3$, the content of CaO, and its coal source (anthracite, bituminous, sub-bituminous, or lignite). FA that contains a $\text{SiO}_2+\text{Al}_2\text{O}_3+\text{Fe}_2\text{O}_3$ of 50 % and CaO content greater than 18 % is classified as Class C and its source is lignite or sub-bituminous coal while FA that contains a $\text{SiO}_2+\text{Al}_2\text{O}_3+\text{Fe}_2\text{O}_3$ content of 50 %, and CaO content less than 18 % is classified as Class F and its source is anthracite or bituminous coal [3]. Class F has pozzolanic properties while Class C has both pozzolanic and self-cementing properties due to higher levels of CaO which makes it harden and gain strength, with/without activators such as lime ($\text{Ca}(\text{OH})_2$), OPC, or gypsum, in the presence of water. The fly ash used in the present study is classified as Class F because the CaO content is less than 18% and the $\text{SiO}_2+\text{Al}_2\text{O}_3+\text{Fe}_2\text{O}_3$ content satisfies 50 wt%. The physical and chemical properties of FA depend on the particle size and type of coal utilized and the techniques used in burning the coal [34], [35]. The chemical composition of the Class F fly ash and phosphogypsum used in the present study is suitable for producing cementitious products and closely agrees with the chemical composition ranges from studies by Authors [35], [36], [37], [38], [8], and [14]. Overall, FA is rich in amorphous silica (range 43 – 67%) and alumina (range 22 – 30%) while PG is rich in calcium oxide (range 31 – 44%) and sulfate (range 30 – 55%). Due to the favourable chemical composition of the FA and PG and their suitability for use as supplementary construction materials, the disposal of FA and PG in landfills can be minimized thereby protecting the environment. The chemical elements present in FA and PG i.e., Al^{3+} , Si^{4+} , Fe^{2+} , Ca^{2+} , instead of being leached into the ground can be incorporated into cement for construction applications.

3.2. pH

The pH is crucial in determining the surface charge of the FA and the ionization and speciation degree of the elements [14]. Figure 1 shows that FA has a pH of 10.63 making it alkaline, PG has a pH of 4.55 making it acidic due to the residual acid from the production process, and 70FA:30PG has a pH of 8.22 making it alkaline. Overall, the FA+PG mixtures can be grouped into strongly alkaline (pH 9 – 11), mildly alkaline (pH 8 – 9), and acidic (pH 4 – 7). Gitari et al. [14] observed that FA mostly develops an alkaline pH shortly after mixing with water due to the large amounts of soluble basic oxides rather than acid phases. pH plays a role in the chemical processes required to produce cement-based composites which mostly occur in alkaline conditions (pH 8 – 13) to allow for pozzolanic reactions and cementitious bonding [39]. Figure 1 shows the linear regression between pH and the FA+PG mixtures giving an R^2 of 0.9401 implying a strong relationship between the pH and PG inclusion i.e., an increase in the amount of PG in the binary mix leads to a decrease in the pH and vice versa.

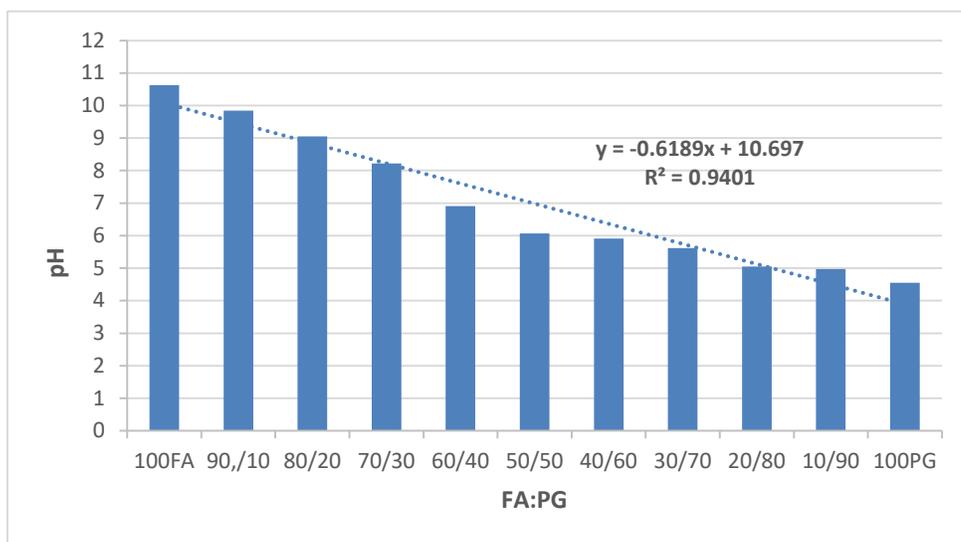


Figure 1. Relationship between pH and FA+PG mixtures

3.3. Leaching

Table II and Figure 2 show the leachate concentrations for FA, PG, and FA+PG mixture in comparison to the regulatory threshold limits. The data is in mg/L expressed as the sample's mean (μ) \pm standard deviation (SD). The error bars show minimal SD of the measured and expected data. The concentration of Mercury (Hg) in the FA, PG, and FA+PG was 0.008 mg/L, 0.004 mg/L, and 0.002 mg/L, respectively, with an SD of 0 mg/L. The measured concentrations of Hg in the FA, PG, and FA+PG mixture comply with the US EPA TC limits and DWAF target water quality (TWR) of South Africa. The Hg regulatory limit is not available in the US EPA land disposal regulation (LDR) limits and EU waste acceptance at landfills. The study observed an immobilization trend in Hg for the binary form FA+PG having the lowest concentration of 0.002 mg/L compared to the unary forms FA and PG representing a 75 % and 50 % decrease in Hg, respectively. Similarly, the leachate concentrations in Pb, Cu, Cd, Cr, Zn, and Ni (ranging from 0.0 to 0.36 ± 0.04 mg/L) were below the US EPA TC, DWAF TWR, US EPA LDR, and EU waste acceptance limits. The low concentration levels of Pb, Cu, and Zn can be due to the formation and adsorption of the negatively charged FA particle surfaces to the positively charged heavy metal ions facilitated by electrostatic forces making them not easily soluble. The leaching of Cr is amphoteric, and its concentration increases based on the dissolution of oxyhydroxide complexes at a high or low pH [45]. However, the concentration of As and Se in PG (0.02 ± 0 mg/L; 0.0 ± 0 mg/L) and FA+PG (0.01 ± 0 mg/L; 0.0 ± 0 mg/L) were within the regulatory limits except in FA where the As (0.15 ± 0 mg/L) and Se (0.07 ± 0 mg/L) were higher than the DWAF TWR limit which can be attributed to their high solubility and mobility in alkaline solutions agreeing with assertions by [46], [47], [48].

The concentration of P in FA (0.42 ± 0.1 mg/L), in PG (2.47 ± 0.7 mg/L), and FA+PG (0.28 ± 0.2 mg/L) was below the US EPA LDR limit. The concentration of Fe in PG (0.03 ± 0.01 mg/L) and FA+PG composite (0.24 ± 0.01 mg/L) were within the DWAF TWR limit except in FA where the Fe concentration (5.51 ± 0.4 mg/L) was higher than DWAF TWR. The concentration of Mn in PG (0.0 ± 0 mg/L) and FA+PG (0.02 ± 0.01 mg/L) was within the DWAF TWR limit except in FA where the Fe concentration (5.51 ± 0.4 mg/L) was higher than DWAF TWR. Gitari et al. [14] observed that the lower Fe and Mn concentrations in FA can be attributed to the adsorption of their metal oxyhydroxides to the negatively charged surface silicate grains of FA in alkaline solutions. Therefore, the lower concentrations of Fe and Mn in the FA+PG composite can be attributed to the precipitation of Mn, Fe, and Cr oxyhydroxides forming distinct granules of mineral oxides. The disintegration of the oxyhydroxides happens following the liberation of Mn and Fe into the solution [14].

Table II. Leachate concentrations in FA, PG, and FA+PG mixture in comparison with different regulatory limits (Data in mg/L, mean \pm SD)

Element	Hg	Pb	Cu	Cd	Cr	Zn	Ni	As	P	Co	Fe	Se	Mn
FA	0.008 \pm 0	0.13 \pm 0.01	0.06 \pm 0.02	0.01 \pm 0	0.04 \pm 0	0.04 \pm 0.01	0.36 \pm 0	0.15 \pm 0	0.42 \pm 0.1	0.1 \pm 0	5.51 \pm 0.4	0.07 \pm 0	0.33 \pm 0.1
PG	0.004 \pm 0	0.02 \pm 0.03	0.03 \pm 0.01	0.01 \pm 0	0.03 \pm 0	0.02 \pm 0.04	0.0 \pm 0	0.02 \pm 0	2.47 \pm 0.7	0.0 \pm 0	0.03 \pm 0.01	0.0 \pm 0	0.0 \pm 0
FA+PG	0.002 \pm 0	0.03 \pm 0.01	0.02 \pm 0.01	0.01 \pm 0	0.01 \pm 0	0.01 \pm 0.02	0.1 \pm 0	0.01 \pm 0	0.28 \pm 0.2	0.01 \pm 0	0.24 \pm 0.01	0.0 \pm 0	0.02 \pm 0.01
US EPA TC Limit	0.2	5.0	n.a	1.0	5.0	2.0	n.a	5.0	n.a	n.a	n.a	1.0	n.a
LDR Limit	n.a*	0.75	n.a	0.11	0.6	4.3	11	5.0	6.0	n.a	n.a	5.7	n.a
EU Limit	n.a	0.5	2	0.04	0.5	4	0.4	0.5	n.a	n.a	n.a	0.1	n.a
DWAF TWR	0.05	0.1	0.1	0.05	0.05	0.1	n.a	0.02	n.a	n.a	0.3	0.02	0.046

n.a * (not available)

Overall, the heavy metal concentrations in the binary FA+PG mixture decreased from their unary concentrations which can be attributed to the immobilization of the ions within the composite network. This assertion agrees with [49] who observed a reduction of heavy metal concentrations in the phosphogypsum-based cemented paste backfill. In usages that depend on the cementitious nature of FA and PG, heavy metal leaching is minimal due to their immobilization by the cement matrix. This assertion agrees with the authors [50], [51], [52], [53], [54] who observed that the matrices of cementitious composites are capable of immobilizing heavy metals and toxic wastes.

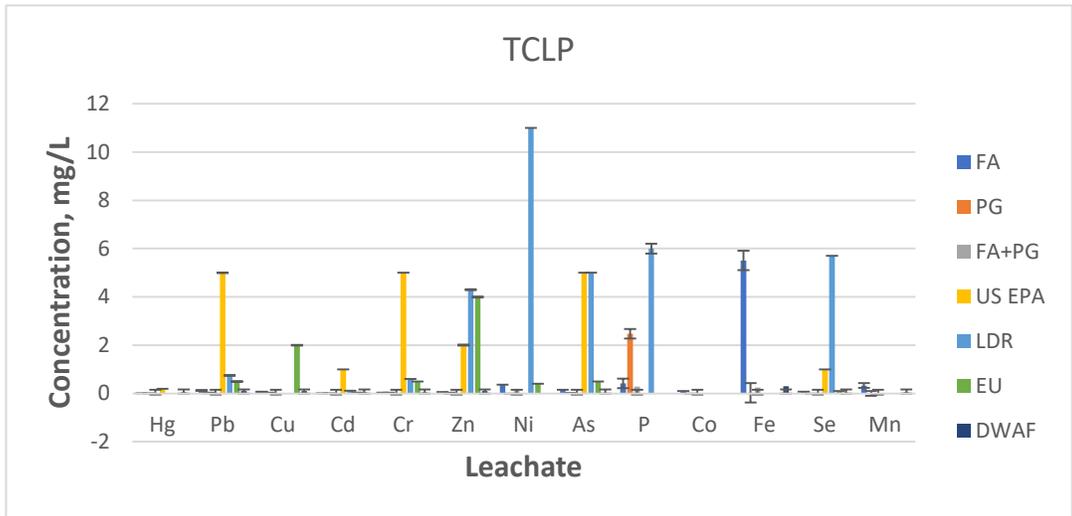


Figure 2. The leachate concentrations in FA, PG, and FA+PG compared to the regulatory limits.

Prolonged landfilling of the FA and PG has the potential to release dust and cumulative amounts of leachates of heavy metals and/or trace elements into the environment. For example, the As and Se in the stockpiled FA will adsorb to soil and groundwater over time via an inner-sphere adsorption mechanism, forming bidentate surface complexes [55]. However, when the unary FA and PG are made into a binary FA+PG mixture, the leachates are reduced and immobilized within the dense compact structure through adsorption reactions. The rate of adsorption of As escalates in Fe oxides and its mobility depends on the waste chemistry and pH [56]. Therefore, it is imperative to promote the solidification of FA and PG not only to adsorb and/or immobilize these toxic elements but also to minimize landfills, increase usage in the construction industry, support the circular economy, and advance SDGs 9, 11, 12, and 13. It is evident from the foregoing that the reuse of FA and PG is beneficial due to the immobilization of heavy metals compared to disposing of them in landfills which brings about undesirable leaching effects over time at the disposal area. Furthermore, the chemical elements of economic interest (e.g., Zn, Cu, Fe, P) can be recovered from the leaching of FA and PG. Leaching is an important technique for recycling valuable metals [57]. The recycling of FA and PG can help reduce landfill exposure to weathering processes, eliminate waste disposal costs, and provide alternative construction materials for sustainable infrastructural development.

To validate the results obtained in the present study and predict potential pollution from leaching at a global scale, a comparison of analyses was made to leachate concentrations found in other countries (Table III). The present study’s findings on FA, PG, and FA+PG leachate concentrations were lower and/or within those concentrations reported for Spain, India, China, USA, Australia, United Kingdom, Turkey, Jordan, and Morocco. Since the heavy metal and/or trace element concentrations are within the regulatory limits, the FA, PG, and FA+PG used in the present study can be considered non-hazardous and appropriate for use in construction applications. The US EPA ruling in 2014 classified coal fly ash as non-hazardous but advised the importance of monitoring the heavy metal concentrations to comply with the US EPA regulatory limits [35], [58]. This advice agrees with [14] who observed that high heavy metal concentrations leach out of FA due to time series variation of the pH negatively affecting the environment or groundwater if the disposal

area is not properly designed. The problems of disposing of FA and PG in dams or landfills lead to the leaching of heavy metals and/or trace elements into water bodies and soils creating toxic effects on living organisms through bioaccumulation [21], [56]. For example, Hg causes nervous and circulatory system disorders, and aquatic failure, Cr causes diarrhoea, nausea, and headache; Cu causes anaemia, insomnia, liver damage, and soil enzymatic inhibition; and Pb causes kidney failure, infant brain damage, muscle impairment. Furthermore, these landfills require pretreatment before disposal and continued monitoring thereby incurring variable costs [59]. Therefore, promoting the circularity and usage of FA and PG in the construction industry is imperative to minimize environmental pollution from landfill leachates.

Table III. FA and PG leachate concentrations reported worldwide (Data in mg/L)

Element	Country	Hg	Pb	Cu	Cd	Cr	Zn	Ni	As	P	Co	Fe	Se	Mn
FA [16]	Spain	0.008	0.035	0.046	0.006	1.375	0.943	0.015	0.146				2.617	
FA [60]	India		0.636		0.086	0.079	0.055	12.4	0.346		0.134	14.08	0.6066	0.44
FA [15]	China		4.40	4.61	0.51	1.23			7.73					
FA [61]	USA	0.15	19	13.5	0.03	24.8	40.4	23.0	74.6		13.5	13333	0.2	102
FA [62]	Australia		48	28.10	0.25	18		10.5	6.58	648	5.6	48300	2.48	899
FA [14]	South Africa			0.0033		0.0026	0.0046	0.0067			0.0011	0.18	0.00037	0.19
FA [13]	Turkey		0.37	0.01							0.06	0.80		0.13
FA [63]	UK		<0.01	0.234	0.01	0.013	0.86	0.315				0.060		1.00
PG [31]	Spain		0.002		0.010	0.005	0.026	0.011	0.002				0.004	
PG [64]	India		0.0112	0.012	0.0035	0.032	0.005		<<0.01			0.24		
PG [32]	China		0.0146	0.1921	0.0114	0.0138	1.2245			128.9969	0.0676			
PG [65]	USA		1	8	7		9	2	42		2	670		15
PG [65]	Jordan			0.02	0.02	0.08	0.09	0.07	0.18		0.03	2.10	0.21	0.22
PG [17]	Morocco		1.92	7.1	1.69	4.23			1.02					
PG [66]	Morocco		<0.60	0.19	<2	0.56	2.05	0.3	0.56		1		0.26	

4. Conclusion

This study comparatively assessed the leaching properties of FA and PG disposed of in landfills at Eskom Duvha Power Station in Mpumalanga and Foskor (pty) Ltd in Phalaborwa, South Africa. It was found that the FA and PG are non-hazardous since the leachate concentrations of Hg, Pb, Cu, Cd, Zn, As, and Se are within the US EPA TC, US EPA land disposal regulation, and EU waste acceptance at landfills. Compared to the unary forms of FA and PG, the binary mixture of FA+PG had the lowest leachate concentrations. The solubility and mobility of As and Se depend on the alkaline concentration of the solution. The rate of adsorption of As escalates in Fe oxides and its mobility depends on the waste chemistry and pH. The PG threshold limit of 30 wt% put the FA+PG mixture within the alkaline ranges (pH 8 - 13) suitable for the promotion of pozzolanic reaction and cementitious bonding as construction materials. PG is acidic such that if added at proportions greater than 30 wt% can make the FA+PG mixture acidic thereby affecting physicochemical properties. The leachate concentrations in FA, PG, and FA+PG comply with the regulatory limits and are thus considered non-hazardous and suitable for use in construction. The chemical elements present in FA and PG i.e., Al³⁺, Si⁴⁺, Fe²⁺, Ca²⁺, and S²⁻, instead of being leached into the ground can be incorporated into cement for construction applications. Furthermore, the chemical elements of economic interest (e.g., Zn, Cu, Fe, P) can be recovered from the leaching of FA and PG. The recycling of FA and PG can help reduce landfill exposure to weathering processes, eliminate waste disposal costs, and provide alternative construction materials for sustainable infrastructural development.

5. Limitations and further research

FA and PG were collected within South Africa. Leachate concentrations and chemical compositions may vary in different environments. Therefore, further research should extend to different local and global sources. Further studies

should also investigate the impact of electrical conductivity and temperature variation on the leaching properties of FA and PG, develop chemical prediction models for leachates in FA and PG, and develop cost-effective treatment and extraction methods of elements of economic interest from FA and PG.

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