Beneficiation of Barberton gold mine tailings: The effect of fly ash on the mineralogy and micrograph

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In this study, the chemical composition and morphology of fly ash stabilised gold mine tailings was examined, to evaluate its possible application for building and construction industry. Cyanide treated gold mine tailings (treated from the source), where gold has already been recovered was used in this study. The tailings were stabilised with fly ash, to improve its properties. Specimen at the maximum dry density and optimum moisture content were prepared and cured at low and elevated temperatures of 40 °C for 7 days and 80 °C for 4 days, respectively. The effect of stabilizing gold mine tailings using fly-ash at different tailings to fly-ash ratios of 80:20, 70:30, 60:40, 50:50 and 40:60 were studied. The obtained results showed that at the elevated temperature, hydration products that have a great potential to enhance the strength of the material were formed. Fly-ash stabilized gold tailings and increased the relative proportion of calcium oxide and alumina that contribute to strength enhancement.

Keywords: Beneficiation, stabilisation, fly-ash, gold tailings

INTRODUCTION

South Africa has for many years remained the world leader in mining, with the abundant mineral resources such as precious metals; gold and platinum group metals and copper. The production of gold has made a significant impact on the city-region's natural landscapes and has played an influential role in the shaping of Johannesburg and other 'Rand towns' (Bobbins and Trangos, 2018). In 1890 other types of mining operations, such as coal, were discovered in eMalahleni, which are now primary to the economy of the region (Bobbins and Trangos, 2018). These coal deposits have made a significant contribution to the South African economy and are considered secondary to the city-region's gold exploits (Bobbins and Trangos, 2018). During the production of gold from the ore, gold and waste by-products are produced, where the gold is sold and the waste tailings are sent to the dumps or landfills. Landfills pose an environmental hazard, leading to contamination of valuable resources such as water and soil. The acid in the tailings contaminates the drinking water and pollute the soil and ground water (Phetla et al., 2012).

For many years, South Africa has been facing a major challenge related to acid mine drainage (AMD), due to these gold tailings. AMD is formed when there is oxidation of sulphide minerals in tailings due to exposure to moisture and oxygen (Nengovhela et al., 2006). More than 5t of solid and liquid waste by-products is generated in the gold production process (mining and milling processes) to obtain about 10g of gold, (Ripley et al., 1982). Only a small percentage of these waste by-products is currently used for backfilling or construction purposes in South Africa and the majority of the material is disposed into landfills. It was acknowledged by the Department of Water Affairs and Forestry (2001) that waste generated by gold mining is the largest single contributor to waste and pollution, and has accounted for 47% of the total mineral waste in South Africa. Of the waste, the tailings dams alone account for some 6 billion tonnes that resides in 270 dams spanning a collective area of some 400 km^2 (Oelofse et al., 2007) across the Witwatersrand. These challenges in connection with the increasing landfill costs, the stricter enforcement of environmental legislation and implementation on waste disposal, calls for innovative methods for the utilisation and beneficiation of gold tailings for other applications (Malatse & Ndlovu, 2015).

In searching for solutions to utilise gold mine tailings, numerous researchers have explored the applications of these waste by-products for building and construction. Due to the chemical characteristics and geotechnical properties, most of the gold mine tailings require stabilisation before the material can be used for any application. Rachman et al. (2018), investigated the stabilization and solidification of tailings from a traditional gold mine using Portland cement. The result obtained showed that, the optimum ratio to mix Portland cement and tailings was 10:90, with compression test of 257 ton/m^2 and Toxicity Characteristic Leaching Procedure (TCLP) test was 0.0069 mg/L. The compression test results were in accordance to US EPA Standard quality of 35 ton/m^2. TCLP test results met the standard of Indonesian Government Regulation No. 101 Year 2014 of 0.05 mg/L (Rachman et al., 2018). In studying the potential for reuse of gold mine tailings as secondary construction materials and Phytoremediation, Mapinduzi et al. (2016), discovered that, the gold tailings used can be classified as alkaline, non-plastic silt materials with low organic (< 2.5%) and nitrogen (<1%) contents; typical of hard rock tailings. The tailings had a pH varying from 7.2±0.31 to 7.51±0.11, typical for soils with large amounts of Ca^{2+} and Mg^{2+}. Low CEC values (< 10 meq/100g) and conversely low levels of organic content (< 2.5%), and low nutrients content (N < 16.5 mg/kg and P < 1.0 mg/kg), indicating that tailings required amendments with Terminal Electron Acceptors (TEA) in the form of fertilizers to enhance its capacity to hold any nutrients.

Jia et al. (2012), conducted a study on the use of residual products as additive in tailings paste for the mitigation of ARD: Effect of green liquid dregs and fly ash addition on geotechnical stabilization of tailings. The study focuses on the effect of green liquor dregs (GLD) and fly ash (FA) on geotechnical stabilization of tailings, using shear strength test. Addition of FA to the paste greatly improved shear strength up to 2-3 times higher. Moreover, the fly ash hardening process effectively reinforced the strength of GLD amended tailings paste. Longer curing period (3 months) for specimens lead to up to 2-3 folds higher strength compared to that of 1 month. Hydraulic conductivity was reduced as a result of GLD and FA addition, since the porosity of the tailings decreased. Mine tailings have been used as a substitute construction material, to recycle waste products, for the production of hollow blocks and bricks as masonry mortar. Gopez (2015), investigated the use of Philex copper-gold mine tailings (PCGMT) in roller compacted concrete (RCC) production. Comprehensive experiments were conducted to investigate the comparative compressive strengths of RCC containing Porac sand (RCCPS) and RCC with copper-gold mine tailings (RCCCGMT), and the durability of RCC with copper-gold mine tailings and fly-ash. The compressive strength attained at 28 days by RCCPS and RCCCGMT ranges from 17 to 37 MPa and 17 to 28 MPa, respectively. The obtained values showed that the latter required greater amount of cement and the mixtures with compressive strength values greater than 25 MPa are acceptable for concrete pavement use. The durability of RCCCGMTFA with medium cement content was evaluated by subjecting the specimens to an alternate wetting and drying cycles. After 15 cycles, a remaining strength of 18.7 MPa was obtained which indicated that it could endure stresses.

In 2015, Kunt et al., investigated the utilization of Bergama gold tailings as an additive in the mortar, in the study the effects of the gold tailings on the compressive strength properties of Portland cement were investigated. Gold tailings with different ratios of 5, 10, 15, 20, and 25 % were added as an additive in the mortar with varying ratios of cement and sand. According to results, it was concluded that the tailings are eligible for mortar aggregate and the optimum ratio of gold tailings is 5%. The micropore structure of cement-stabilized gold mine tailings was studied by Lee and Shang (2018). The samples were prepared with different fractions of tailings and cement, cured at elevated temperatures, and activated addition on chemical ($CaCl_2$). It was observed that all mixed samples exhibit a mono-modal pore size distribution, indicating that the cement-stabilized tailings are characterized by a single-porosity structure. The results also showed that the higher fraction of tailings and cement leads to a dense and finer pore structure. In this study gold mine tailings was stabilised with fly ash at different ratios, the mineralogical and micrograph development were studied. Based on the properties of the stabilised material, recommendations are also made for the applications of gold tailings in building/constructions.

EXPERIMENTAL

The treated gold tailings were collected from Pan African Resources Mine in Barberton, Mpumalanga.

The sample were dried in the oven at a temperature of 50°C. The dried material was characterized for elemental analysis using X-ray fluorescene (XRF), mineralogy using X-ray diffraction (XRD) and morphology by Scanning Electron Microscope (SEM). Sieve analysis and Malvern analyser were conducted to study the particle size distribution. The standard compaction tests were conducted to determine the maximum dry density (MDD) and optimum moisture content (OMC) of the tailings, following ASTM D698. After obtaining the MDD and OMC of the material, the specimen were cast in a 100mm x 100mm moulds and cured for 4 days at elevated temperatures of 80 ^{o}C and 7 days at 25 ^{o}C temperature. The gold tailings were then stabilised with fly ash, by varying the ratios of tailings to fly-ash ratios by 80:20, 70:30, 60:40, 50:50 and 40:60. After stabilisation the specimen were cast and cured at 80 ^{o}C, thereafter the elemental, mineralogy and morphology of the specimen were studies. A thorough study was conducted on the mineralogy of the composites to know the new hydration products formed.

RESULTS AND DISCUSSION

Elemental analysis of un-stabilised gold tailings

Table 1 shows the XRF analysis of gold tailings of Barberton tailings before stabilisation.

Table 1. XRF analysis of gold tailings and fly ash (wt %)

Material	SiO_2	AlO_3	MgO	CaO	Fe_2O_3
Tailings Barberton	48.10	11.3	4.6	9.2	14.9
Fly ash	47.90	28.00	-	5.13	4.83

From the elemental composition shown in Table 1, it is evident that the major oxides contained in tailings from Barberton, were SiO_2, Fe_2O_3 and Al_2O_3, and CaO being the fourth abundant constituent. The tailings have similar composition as the results reported by Rudzani et al. (2017), where these four oxides were identified in gold tailings from Sabie's Pilgrim Rest. In their study to characterise the gold tailings hey found that the material was laden with SiO_2, Fe_2O_3 and Al_2O_3 and CaO, as the predominant constituents. Fly ash consisted of constituent such as silica, calcium oxide, and in minimal relative proportion ferrous iron and titanium oxide, the results similar to what Mashifana et.al (2018), reported.
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Mineralogical analysis of un-stabilised gold tailings

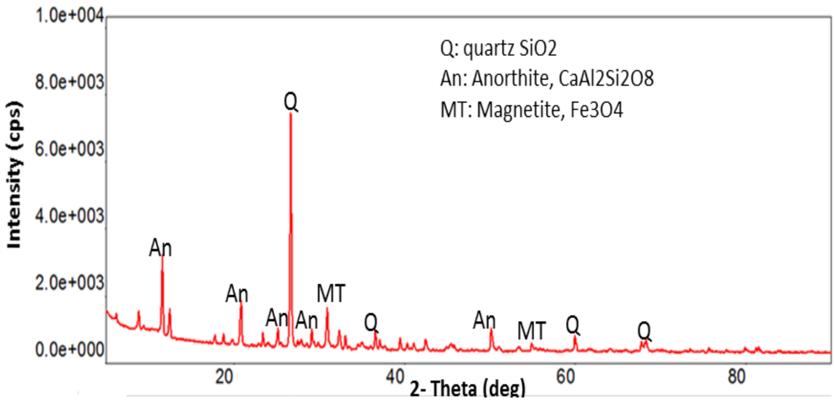

Figure 1. XRD analysis of un-stabilised gold tailings.

The mine tailings are mainly crystalline materials consisting of quartz, anorthite and magnetite, Figure 1. The mineralogy agrees with the elemental composition presented in Table 1. Anorthite is a dominant mineral at 63.39%, followed by 35.98% quartz and 0.68% magnitite. The availability of quartz may be attributed to its dominance in the original tailings as well as to its high resistance to physical and chemical weathering and dissolution (Rudzani et al. 2017). The main host rock in Barberton area consists of high quantity of quartz up to 50%, which also contributed to the silicate content (Altigani et al. 2016).

Maximum dry density and moisture content of un-stabilised gold tailings
The MDD and OM of gold tailings are presented in Figure 2.

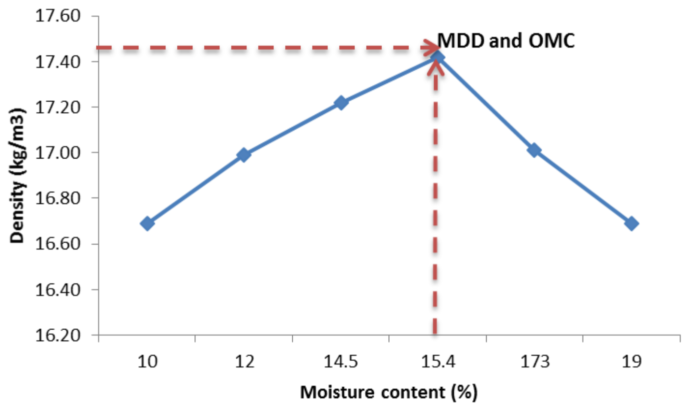

Figure 2. MDD and OMC of gold tailings.

The MDD was found to be 1742 kg/m^3. The increase in dry density continues until it reached a point, where water starts occupying the space that could have been occupied by the soil grains this is the MDD and the OMC was found to be 15.4%. As the material was compacted the voids were reduced and this resulted in increase of the dry unit weight or dry density. Initially as the moisture content increased so did the dry unit weight. However, the increase did not occur indefinitely due to the material state approaching the zero air voids line which provides the maximum dry unit weight for given moisture content.

Particle size distribution of un-stabilised gold tailings
The coefficient of gradation (Cu) and uniformity coefficient (C_C) of the tailings was calculated using Equation 1 and Equation 2. The coefficient of gradation, Cu, is a parameter which indicates the range of distribution of grain sizes in a given specimen. If Cu is relatively large, it indicates a well graded soil. If Cu is nearly equal to one, it means that the soil grains are of approximately equal size, and the soil may be referred to as a poorly graded soil (Das, 2002). The parameter Cc is also referred to as the coefficient of curvature, if Cu is greater than 6 and Cc is between 1 and 3, it is considered well graded

$$Cu=\frac{D60}{D10} \quad [1]$$

$$Cc=\frac{(D30)^2}{(D10)(D60)} \quad [2]$$

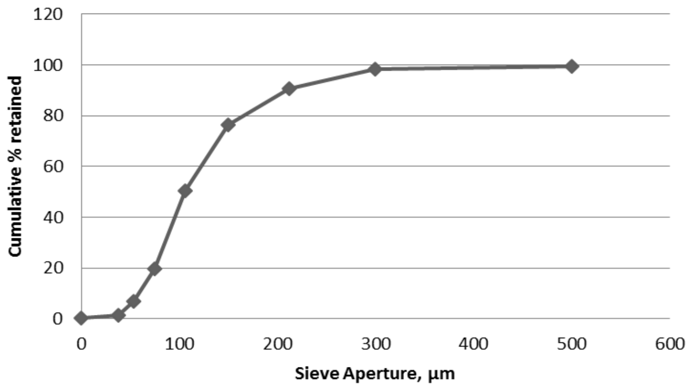

Figure 3.PSD of gold tailings.

Figure 3 presents a particle size distribution curve of gold tailings. The co-efficient of gradation (Cu) for the gold tailings is 2.038 and the uniformity co-efficient (Cc) was 0.78, these values indicate that the tailings was well graded. In 2014, Lee et.al, also found that, gold tailings tend to be well graded and primarily silt-sized particles, classified the material according to Unified Soil Classification system (USCS).

Density and pH of gold tailings

A pycnometer was used to determine the density of gold tailings. The density of the material was measured to be 2.80 g/cm^3. Lee et al. (2014) reported a density of 3.28 g/cm^3 in gold mine Musselwhite which is greater than of most soil, while Zhang et al. (2011) reported a density of 2.83 g/cm^3 which agrees with the density of the gold mine tailings used for this study. The pH of the gold tailings was measured to be 8.94, which indicate that the material is alkaline and the tailings lead to a non-acidic discharge. The pH values of 3.25-6.28 for gold tailings are mostly recorded in South Africa (Fashola et al. 2016). These are normally the tailings where gold has not been further recovered. The pH of 7.35 was reported in gold tailings from Iran (Rafiei et al. 2010), while Davies (2002) reported pH values of 3.48-8.12 in India. Malatse and Ndlovu (2015), reported pH values of 4-7 from the Witwatersrand gold tailing in South Africa, indicating an acidic material. Gold mine tailings are either acidic or alkaline depending on the processes utilised for gold recovery. This also indicates that, for Baberton mine, further recovery of gold contained in gold tailings by using cyanide as a leaching reagent, resulted into an alkaline material.

Elemental composition of tailings after curing at low and elevated temperature

Table 2. XRF analysis of gold tailings after curing (wt %)

Material	SiO_2	AlO_3	MgO	CaO	Fe_2O_3
Tailings Baberton	48.10	11.30	4.60	9.20	14.90
7 Days Curing (25 °C)	45.55	13.50	4.48	9.47	14.29
4 Days curing (80 °C)	49.95	14.14	4.70	8.44	14.96

An increase in curing temperature from 25 °C to 80 °C significantly increased the relative proportion of silica and aluminium, the constituents that promote a pozzolanic reaction and enhance the strength of the material, Table 2. Ahmari and Zhang (2011) also stated that increasing the curing temperature helps accelerate the dissolution of silica and alumina species which exhibit significant pozzolanic reaction.

The effect of curing temperature on the mineralogy of gold tailings

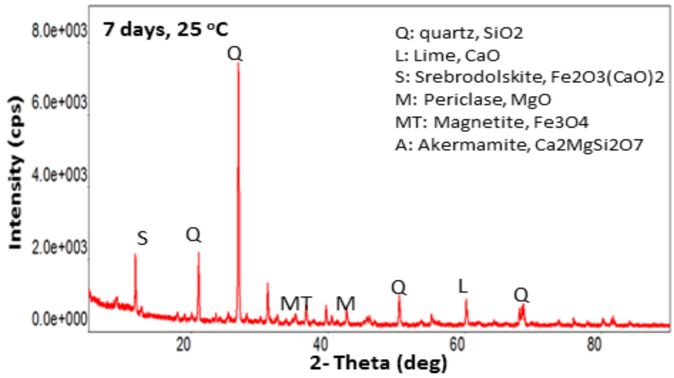

Figure 4. XRD analysis of gold tailings cured at lower temperature.

The mineralogy of gold tailings cured at lower temperature of 25 °C is depicted in Figure 4. The predominant phases are 78.23% SiO_2, 5.78% Fe_3O_4 , 5.71% $Ca_2Mg(Si_2O_7)$, 3.43% MgO, 3.69% CaO, 3.17% $Fe_2O_3(C_aO)_2$.

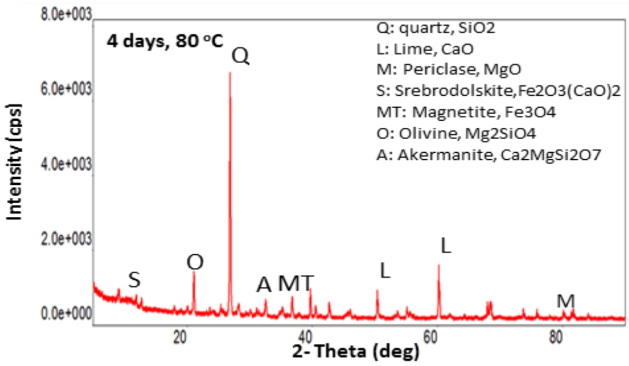

Figure 5. XRD analysis of gold tailings cured at elevated temperature.

The phases detected in the gold tailings cured at elevated temperature of 80 ˚C are that of 92.12% SiO_2, 5.70% $Fe_2O_3(CaO)_2$, 0.83% CaO, 0.37% Fe_3O_4 and 0.34% $Ca_2Mg(Si_2O_7)$. The phase of the gold mine tailings detected correspond with the XRF results in Table 2, the quartz that is associated with the silica, wustite mineral form of iron, olivine the magnesium iron silicate form, lime (CaO), Periclase (MgO) and iron diiron oxide. Curing at elevated temperatures formed lime as a new hydration product, a constituent that also increases the strength of material.

Table 3. Hydration products formed during curing at lower and elevated temperatures

Minerals	Raw gold tailings	7 days curing @25˚C	4 days curing @80˚C
SiO_2	35.93	78.23	92.19
MgO	-	3.43	0.07
M_2SiO_4	-	-	0.5
$Ca_{2.8}.Fe_{.8.7}.Al_{.1.2}.Si_{0.8}O_{20}$	63.39	-	-
CaO	-	3.69	0.83
$Fe_2O_3(CaO)_2$	-	3.17	5.70
$Ca_2Mg(Si_2O_7)$	-	5.70	0.34
Fe_3O_4	0.68	5.78	0.37

From the results presented in Table 3, comparing the gold tailings and the hydration products formed after curing, it is evident that silica increased significantly in the final product, from 35.93% to 78.23 at lower temperature and 92.19 % at elevated temperature. New hydration products that were not initially in the gold tailings were formed. The significant increment of silica and formation of lime and their chemical reaction in water forms a pozzolanic reaction (Dodson, 1990).

Stabilisation of gold tailings with fly ash

Effect of fly ash concentration on the elemental composition

The gold tailings were stabilised with fly ash at different ratios. The following mix designs were prepared, 80:20 Tailings: Fly ash, 70:30 Tailings: Fly ash, 60:40 Tailings: Fly ash, 50:50 Tailings: Fly ash and 40:60 Tailings: Fly ash. The obtained results for the elemental analysis (major oxides) are presented in Table 4.

Table 4. XRF analysis of fly ash stabilised gold tailings (wt %)

Material	SiO_2	AlO_3	MgO	CaO	Fe_2O_3
Tailings Barberton	48.10	11.30	4.60	9.20	14.90
80:20 Tailing: Fly-ash	34.42	11.98	3.14	14.29	21.33
70:30 Tailing: Fly-ash	45.41	15.92	3.93	10.30	13.44
60:40 Tailing: Fly-ash	44.28	16.56	3.63	11.36	12.90
50:50 Tailing: Fly-ash	43.68	17.50	3.62	11.69	12.31
40:60 Tailing: Fly-ash	43.37	17.91	3.10	12.15	13.29

Different ratios of fly ash and gold tailings were studies, to investigate its effect on the elemental composition, as presented in Table 4. The addition of fly-ash significantly increased the relative proportion of alumina and calcium oxide in the mixture. Lee et al. (2014) indicated that, as the fly ash content increases both the OMC and MDD decreases. He further explained that this is due to the physiochemical reaction involving cation exchange, agglomeration and flocculation. Lower OMC and MDD results in formation of hydration products and increase the strength of the material because, the tailings become drier and form coarser particle hence the strength increase Lee et al (2014). The relative proportion of silica decreased with all the mix design. Alumina in the final product increased significantly with the mix design of 30 to 60% fly ash. The relative proportion of calcium also showed a significant increase, up to 14.3 wt % with 20% to fly ash addition.

Mineralogical analysis of fly ash stabilised gold tailings

The XRD analysis of fly ash stabilised gold tailings, at different fly ash ratios is presented in Figure 6 and Table 5.

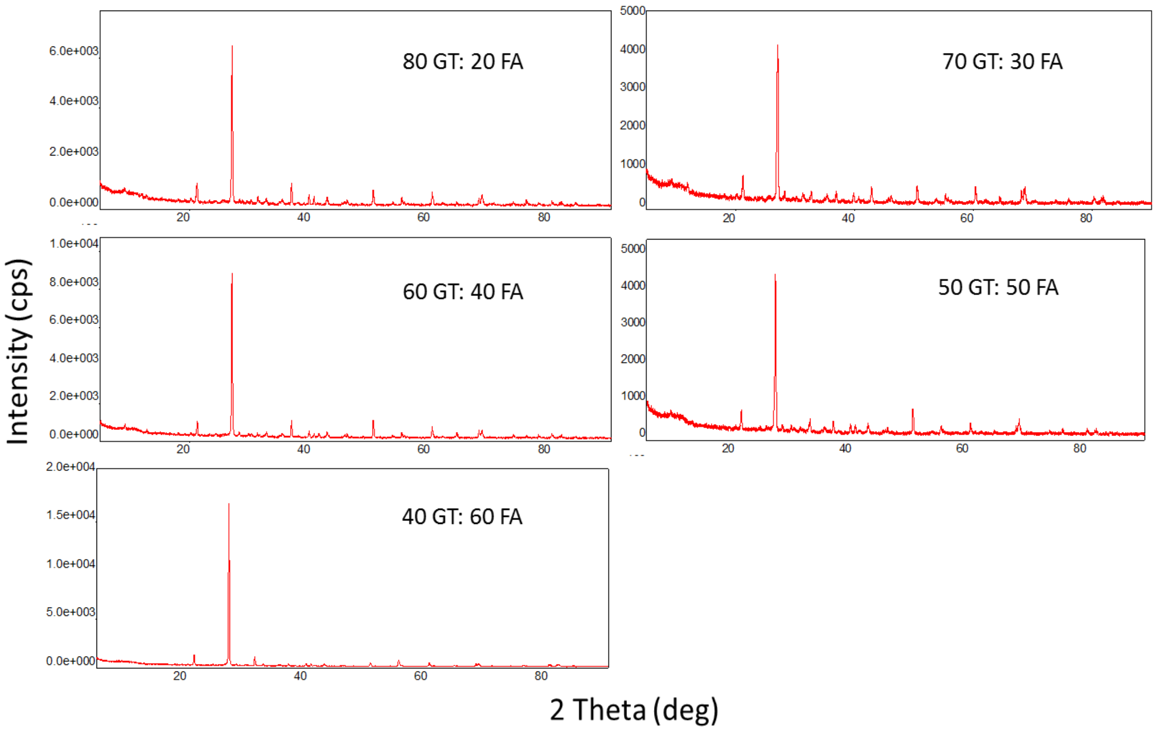

Figure 6. XRD analysis of gold tailings stabilised with fly ash at different ratios.

Table 5. Hydration products formed after fly ash stabilisation of gold tailings

Minerals	Raw gold tailings	80GT:20FA	70GT:30FA	60GT:40FA	50GT:50 FA	40GT:60FA
SiO_2	35.93	77.79	51.90	15.49	82.02	74.84
MgO	-	5.97	-	-	-	-
Al_2O_3	-	10.14	10.58	9.68	2.20	0.49
$Ca_2Fe_2O_5$	-	1.74	0.52		0.16	2.64
Mg_2SiO_4	-	3.31	6.02	73.4	0.75	15.18
$Fe_2O_3(CaO)_2$		-	-	-	-	-
$CaAl_2Si_2O_8$	63.39	-	23.26	-	0.99	0.73
$Ca_2Mg(Si_2O_7)$	-	-	-	-	-	0.19
$Ca_2(SiO_4)$	-	-	7.72	1.59		
FeSi	-	-	-	-	0.86	-
$CaSi_2$	-	-	-	-	13.02	-
Fe_3O_4	0.68	-	-	-	-	

There was a significant increase in the silica content (SiO_2) and Forsterite (Mg_2SiO_4) a mineral associated with silica when the content of fly ash was increased from 20%, 30%, 50% and 60%. This is due to the high silica content available in fly ash. A number of new hydration products were also formed, with fly ash stabilisation. The results also show that lime reacted completely with aluminium and silica, to form new products. When focusing on the pozzolans that plays a role in the development of unconfined compressive strength of the material, fly ash concentration of 20% yielded the best results, as there was a significant increase in both silica and aluminium.

Micrograph development of fly ash stabilised gold tailings

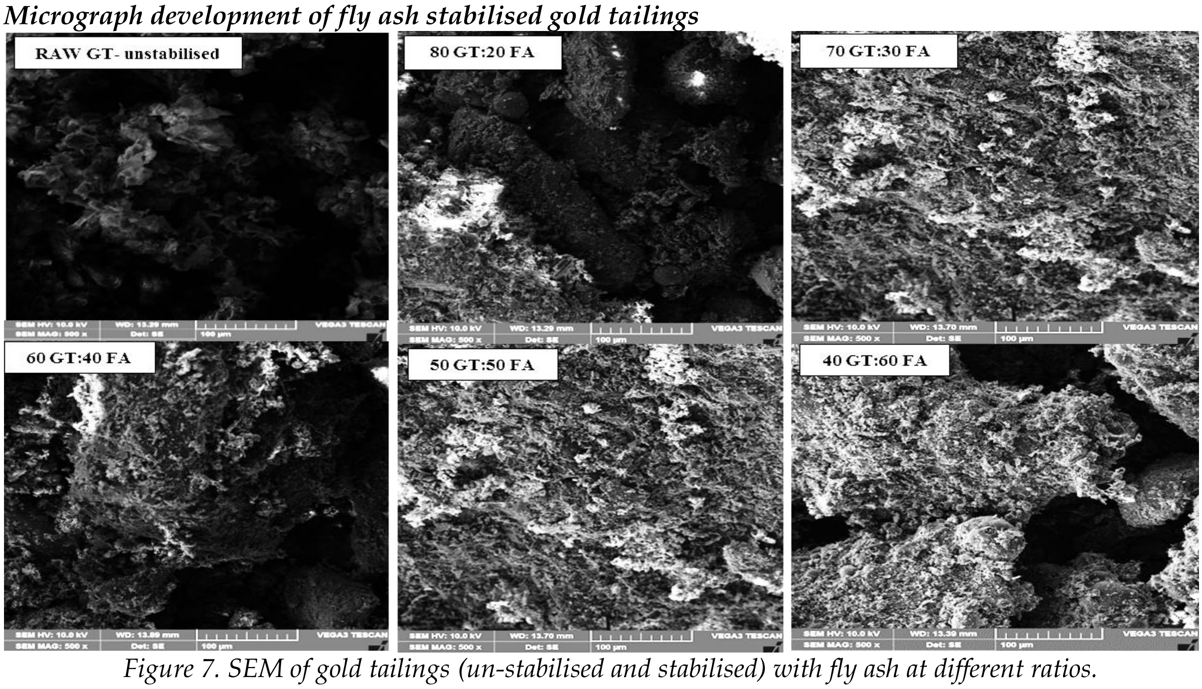

Figure 7. SEM of gold tailings (un-stabilised and stabilised) with fly ash at different ratios.

The micrograph images of fly ash stabilised gold tailings is presented in Figure 7. Substantial finer particles, resembling flakes are observed in the raw gold tailings (un-stabilised). An improvement in the structure of the gold tailings after stabilisation with fly ash is also evident. The new hydration products formed played a significant role in transforming the morphology of gold tailings. More is available in the tailings. More solid particles were formed in the 20% fly ash mix design, a specimen that had the highest content of silica.

CONCLUSIONS

The short term evaluation of fly ash stabilised gold tailings, shows that the tailings that were cured at temperature of 80°C in the oven for 4 days formed hydration products that have a potential to enhance the strength of the material. Fly ash showed to be effective in stabilising Barberton gold tailing by promoting the formation of hydration products that improves the development of unconfined compressive strength in the material. The SEM micrographs of gold tailings was completely transformed by fly ash stabilisation from finer to larger and solid particles.

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