Transformation of Reactive Spoil into Reusable Borrow Materials through Alkaline Waste Valorisation

Juan José Cepriá, Antonio Martinez, Edith Guedella

Technology & Innovation Division, ACCIONA Construction, Valportillo II, 8, 28108 Alcobendas, Spain, juanjose.cepria.pamplona.ext@acciona.com

Abstract Construction sector shows certain lack of expertise on Acid Rock Drainage, ARD, especially where mining activity is insignificant and mining knowledge has not been transposed; consequently, this fact has led to environmental impacts in many civil engineering projects. Nevertheless, construction sector has expertise on maximising materials utilisation by modifying their properties with hydraulic binders. Moreover, recent studies have proposed alternative binders based on waste-products to make projects more sustainable. This work analyses the benefit of the use of wastes commonly used in construction as alternative binders and as alternative treatment for ARD prevention under a Circular Economy approach that could be extended to mine operations, since many of these wastes come from mining-related industries.

Key words Alternative binders, neutralisation, stabilisation, reutilisation, fly ash, ladle furnace slag

Introduction

Civil engineering projects have occasionally to deal with the occurrence of Potentially Acid Forming (PAF) materials as well as alterations of the groundwater table. Road cuts, tunnel boring and earthworks in general, can disturb relevant volumes of PAF rocks and soils. These issues have to be managed according to local regulations and best practices to avoid any environmental risk, which usually comprises treatment and disposal under proper conditions.

In these projects reutilisation of PAF material is normally not foreseen, mainly for environmental reasons, but also because of their poor mechanical properties: lack of bearing capacity, such as in sulphide-enriched coastal plain soils; high swelling potential, due to the precipitation of ettringite/thaumasite; poor durability, due to a quick weathering. Furthermore, the acid leaching from PAF materials can affect the durability of reinforced concrete structures located in their proximity by leaching portlandite out of concrete and inducing cracking in it due to sulphate precipitation within the generated voids. Finally, the acid leachate can prompt corrosion in steel reinforcement bars and anchorages (fig. 1).

***Figure 1** Anchor lines of a concrete retaining wall affected by corrosion (left) and ARD attack to precast concrete elements (right) along the newly-built AP-1 Highway. Guipuzkoa (Spain).*

The ARD phenomenon in civil works is well covered by regulations in those countries where the mining sector is relevant for their economies (i.e. Canada, USA or Australia). In these countries regulations limit the use of borrow materials containing sulphides up to certain threshold values and provide guidance along the project design and implementation. However, they do not usually include valorisation options, although innovative solutions can be accepted after conducting pilot trials (Dear et al. 2014; Ahern et al. 1998). Good examples of the reuse of excavated materials through encapsulation in embankments can be found in Morin et al. 2003. Even so, several accidents have been reported in these countries, such as in the Halifax Airport, Canada (Hicks 2003) or along the I99 Highway, Pennsylvania, USA (Rose and Barns 2008).

These accidents have demonstrated the difficulties and high costs of treating leachates in non-planned and public areas as well as amending these materials with alkaline products to control acid drainage once the problem has appeared.

Soil stabilisation techniques using alkaline binders have been developed in road construction projects along the last 25 years to take advantage of low quality materials excavated along the alignment, so avoiding landfilling and saving in additional borrow materials. The use of alkaline binders has been extended to road bases and subgrades with the aim of reducing their thickness, maintaining their bearing capacity. Consequently, more savings in materials can be achieved.

The main binders used in road construction projects are quick-lime and hydrated lime. Granulated Blast Furnace Slag (GBFS) or coal fly ash are usually combined with these to reduce the economic and environmental costs and to modify properties such as the setting time, hardening process or workability.

The latest research works in this field have been focused on finding alternative hydraulic binders based on alkaline wastes. Several studies have proved the potential of wastes such as paper sludge ash (Segui at al 2012), ladle furnace slag (Manso et al. 2013) and biomass fly ashes (Sarkkinen et al. 2016, Mácsik et al. 2012).

In a parallel way, over the last decade, in the mining sector different alkaline wastes (coal fly ash, different slags, cement kiln dust, green liquor dregs, red mud, construction and demolition waste, etc.) have been successfully used to treat ARD in reactive barriers (Banasiak et al. 2015), in leaching beds (Goetz and Riefler 2014), to directly amend rock waste (Lee et al. 2014) or as component of covers (Mäkitalo et al. 2015). These applications have led to several patents and commercial products, such as Ecotite™, Alkaloam® or Bauxol™.

Unfortunately, little research exits on the possibility of valorising alkaline wastes to amend sulphide bearing spoil and reuse this as borrow materials in infrastructure projects. This research work focuses on the valorisation of low reactive spoil as core material for embankments after being amended through locally available alkaline waste (coal fly ash and ladle furnace slag). The mechanical stabilisation capacity of this technique is assessed to comply with construction requirements and durability commitments, from an integrating perspective.

Materials

The spoil used in this work (RWAS) comes from abandoned stockpiles in the municipality of As Pontes, in the North-West of Spain, generated during the last developments of the local industrial park. The spoil consists of Ordovician alum shales (so-called ampelites) containing muscovite (≈40 % by weight), quartz (≈25 %) chlorite (≈15 %), paragonite (≈15 %), along with minor phases including jarosite, rutile and pyrite. These materials caused the acidification of the river Eume (Blanco 2010), the most serious case of ARD pollution due to civil works, in Spain. Spoil coming from a large road cut along AG-64 highway (see figure 2) in the vicinity of As Pontes, was disposed along the road alignment and rapidly began to leach. This material had been previously identified as PAF at the adjacent As Pontes brown coal mine and had been flooded to prevent ARD, after the largest Spanish mine reclamation project (Aréchaga 1999).

The use of two different binders is explored: fly ash and ladle furnace slag.

Fly ash comes from the nearby As Pontes coal fired power plant. This is a Class C fly ash (high in calcium, see table 1) which was used in the past mine reclamation project to correct soil pH for revegetation of stockpiles. It contains amorphous material (≈ 60 % by weight), quartz (≈ 20 %), some hydraulic phases (merwinite, gehlenite; ≈ 10%) and periclase (≈ 5 %) along with unburnt particles. Its finesse is 18.5% according to EN 451-2.

Ladle furnace slag (LFS) is generated in electric arc furnaces (EAF) to produce steel from scrap and raw materials (lime, dolomite). The first stage of the process produces crude steel and EAF slag, and then the crude steel is refined into high grade steel, generating LFS. It is powdery, strongly alkaline and contains portlandite (≈ 20 %), periclase (≈ 15 %), hydraulic phases (merwinite, larnite; ≈ 30 %), calcio-olivine and quartz. Chemically, LFS consists of CaO, MgO, Fe_2O_3 and SiO_2.

Table 2 shows that the considered fly ash is low in trace elements while LFS contains is high in Cr and Zn as trace elements, compared with regular binders.

Table 1 *Chemical composition; major and minor elements by XRF (%)*

	SiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	TiO_2	P_2O_5	LOI
RWAS	51.07	26.01	7.68	0.037	2.09	0.42	1.30	3.60	1.02	0.14	6.95
FA	37.32	14.57	12.00	0.119	6.12	12.87	0.74	1.01	0.74	0.24	13.36
LFS	14.46	4.26	14.09	2.17	15.69	41.90	0.02	<0.01	0.407	0.14	6.75

Table 2 *Trace elements by IPC-MS and INAA (ppm)*

	As	Ba	Cd	Cr	Cu	Hg	Mo	Ni	Pb	Sb	Se	Zn
RWAS	21	707	<0.5	148	29	<1	<2	13	28	2.0	<3	137
FA	47	1870	<0.5	78	61	<1	5	103	29	2.9	<3	167
LFS	3	482	2.6	932	93	<1	6	26	33	1.2	<3	2450

Figure 2 *As Pontes road cut along AG-64 highway (left) and sampling area at the industrial site (right).*

Methods

A dosage study was carried out to determine the minimum dose rate of fly ash and LFS to be added to spoil to attain its geochemical stabilisation (see table 3), for this reason, Net Acid Generation (NAG) tests and Acid Base Accounting (ABA) characterization were performed. Simultaneously, the neutralisation potentials of the alkaline amendments were calculated for the determined dosages and corrected according to the method employed by the Roads and Traffic Authority of New South Wales (table 3), due to the lack of Spanish standards covering this topic.

Table 3 *Summary of the results of the geochemical characterization (kg CaCO3 equivalent per ton)*

	NAG (actual)	NAG (total)	NAG pH	AP	NP	NAP	Dose Rate (DR)	Corrected DR
RWAS	9.28	14.98	2.50	17.34	2.28	15.06	-	-
FA	-	-	-	-	312	-	4.89 %	9.78 %
LFS	-	-	-	-	925	-	1.65 %	3.30 %

A series of tests were carried out on a set of spoil samples before and after being amended to evaluate the Optimum Moisture Content (OMC), Maximum Dry Density (MDD), CBR, and swelling (table 4). Based on the values obtained for the MDD and OMC parameters, three specimens were prepared by extrusion according to the calculated dose rates in order to conduct column leaching tests.

Table 4 *Summary of the results of the mechanical characterization*

	MDD (ton/m^3)	OMC (%)	CBR index	Swelling (%)
RWAS	2.19	8.1	3.4	3.3
RWAS + 9.78 % FA	2.08	9.1	46.7	0.4
RWAS + 3.30 % LFS	2.15	8.8	23.3	1.1

The specimens had cylindrical shape with a radius of 8.4 cm and heights between 12.0 and 12.4 cm. They were placed in glass leaching column cells with a glass filter covered with a fine 0/0.25 mm quartz sand layer. The gaps existing between the cells and the cylinders were sealed with paraffin wax to make all the water percolate through the specimens.

A solid/liquid ratio equal to 0.2 was selected taking into account the indications contained in common standards and adapting the values to the hydraulic conductivity and water sorption of the specimens. The watering frequency was set to allow the samples to dry up, thus submitting them to wet-dry cycles. The leaching tests had lasted for one year and during this period seven wet-dry cycles were produced. At the end of the tests all leachates were collected and analysed to detect the trace elements included in table 2 , adding in iron concentration. The evaluation of the concentrations of these elements (except for iron) is required by Decision 2003/33/CE which sets the criteria for the acceptance of waste for each landfill class. The testing included also measurements of pH, electric conductivity and alkalinity/acidity release by titration.

Finally, specimens were dried out and ABA tests were carried out to assess the alkalinity and sulphide evolution. Chemical composition (whole rock analysis) and mineralogy were also determined to check the new mineral species. Texture and microtexture were also analysed by means of optical microscopy and SEM.

Table 5 *Summary of the results of the geochemical characterization (kg CaCO3 equivalent per ton) of the specimens after being submitted to leaching tests*

	AP	NP	NAP	Total S	Sulphide S	Paste pH	CaO	MgO
RWAS	5.73	-0.62	6.35	0.42	0.18	4.84	0.45	1.88
RWAS + 9.78 % FA	5.37	14.0	-8.62	0.65	0.17	8.30	1.69	2.28
RWAS + 3.30 % LFS	5.35	24.2	-18.9	0.70	0.17	10.2	1.83	2.38

During the leaching test, the specimens went through an intense leaching process under the typical compaction criteria for embankments. Thus, it could be considered an exigent test, beyond the usual road operating conditions.

Results

Mechanical improvement: The alum shale has poor mechanical properties as borrow material due to its high swell (3.3 %) and low bearing capacity (CBR index of 3.3). Even if other durability-related properties were not tested, it can be observed that usually, shales do not exhibit good durability properties.

After adding a 3.3 % of LFS to the spoil, the CBR index of the treated spoil showed a seven-fold increment with respect to the value correspondent to the untreated spoil. MDD slightly lowered and OMC increased, as expected for limed materials. Swelling reduced to 1.1 % and an important decrease in hydraulic conductivity was also noticed.

Spoil treated with 9.8 % of fly ash exhibited a fourteen-fold increment in its CBR index with respect to the value correspondent to the untreated spoil. MDD, OMC and swelling showed the same trend as in the case of using LFS. The increase obtained in the OMC indicates that it would be possible using wetter materials.

Geochemical behaviour: Trace element concentrations in the collected leachates showed extremely high values for the untreated specimens (blank, fig. 3). These values are clearly above the reference given in "European Decision 75/440 concerning the quality required of surface water intended for the abstraction of drinking water" (red line, fig. 3) and "Law 9/2010 concerning the threshold values for water discharges into Galician coastal inlets" (green, fig. 3).

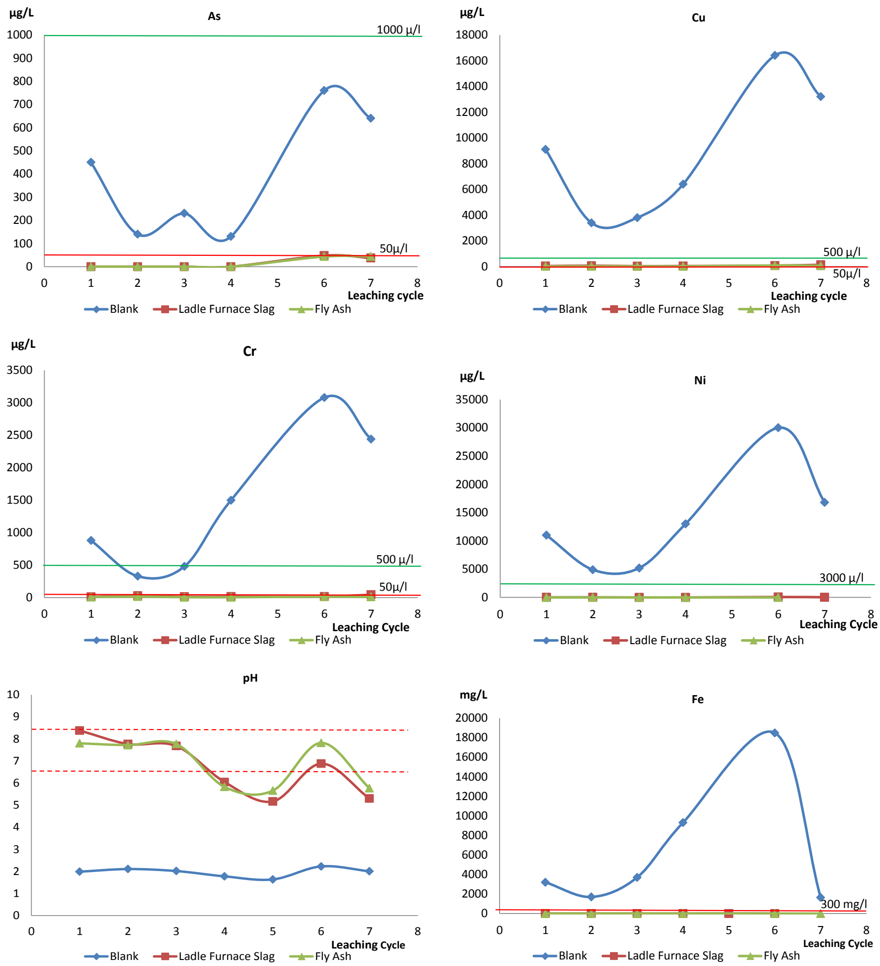

Figure 3 *Evolution of As, Cu, Cr, Ni, Fe and pH along the leaching test. Horizontal lines represent the reference values according to the European Decision 75/440 (red line) and the regional regulation for water discharges, Law 9/2010, Xunta de Galicia (green line).*

For both treated specimens, the concentrations of the dangerous trace elements remained below the aforementioned threshold values. In general, leachates from both treated specimens showed similar concentrations, although the one treated with fly ash exhibit slightly lower values, except for Selenium. Concentrations of all analysed elements in the LFS-treated specimen remained constant and close to till to the last cycle, when a minor rise for all the elements was observed. Leachates from fly ash-treated specimen showed the same minor rise but only in As and Cu. For both treated samples Fe remained close to zero and pH circumneutral up to cycle 4, while from this point on, available alkalinity showed signs of exhaustion, although, they continued having neutralisation capacity till the end of the leaching tests, especially in the case of LFS (table 5).

On the contrary, untreated specimens showed a continuous increase in the concentrations of all analysed elements up to the last cycle, indicating a weak depletion of metal mobility. Values of pH remained constant around 2 and Fe concentration greatly increased up to the last cycle, following the behaviour of the trace elements.

Conclusions

A sulphide bearing alum shale known to have caused ARD pollution in Galician rivers had been treated with two local wastes: fly ash and ladle furnace slag. Results demonstrate that feasible dose rates are able to neutralise acid generation and avoid metal mobility. As a consequence of the proposed treatments, mechanical properties were improved, suggesting that this treated material could be used in road subgrades in compliance with most of the international requirements. These results open the possibility of using *low* reactive spoil in road embankments or general fills in mining operations following a circular economy approach which consists in the use of locally available amendments based on waste and by-products usually associated with mining activities.

Civil works could also benefit from this possibility when it is necessary to manage acid forming materials, adopting the characterization procedure of the mining sector when sectorial regulation and requirements lack.

References

Ahern CR, Stone Y, Blunden B (1998) Acid Sulfate Soils Assessment Guidelines. Acid Sulfate Soil Management Advisory Committee, Wollongbar, NSW. Australia, 63 pp.

Aréchaga F (1999) Considerations upon mine voids based on dump recovering experience. Puentes mine case IMWA Proceedings.

Banasiak L, Indraratna B, Lugg G, Pathirage U, McIntosh G, Rendell N (2015). Permeable reactive barrier rejuvenation by alkaline wastewater. Environmental Geotechnics, 2 (1), 45-55.

Blanco R (2010) Centro de tratamiento de residuos industrials de Galicia. Situación actual y perspectivas de future. XVI Curso de Saude Ambiental, Baiona.

Dear SE, Ahern CR, O'Brien LE, Dobos SK, McElnea AE, Moore NG, Watling KM (2014) Queensland Acid Sulfate Soil Technical Manual: Soil Management Guidelines. Brisbane: Department of Science, Information Technology, Innovation and the Arts, Queensland Government.

Goetz ER, Riefler RG (2014) Performance of steel slag leach beds in acid mine drainage treatment. Chemical Engineering Journal, 240, 579-588.

Hicks SA (2003) Acidic airport drainage, 20 years and $20 million worth of experience. Proceedings of the Sixth International Conference on Acid Rock Drainage, Cairns, Australia, p 14-17.

Lee JK, Shang JQ, Wang H, Zhao C (2014) In-situ study of beneficial utilization of coal fly ash in reactive mine tailings. Journal of environmental management, 135, 73-80.

Mácsik J, Edeskär T, Rogbeck Y, Ribbing C (2012) Stabilization of road structures with fly ash as binder component: through demo projects to full scale use, Ash Utilisation 2012, Stockholm, Sweden.

Mäkitalo M, Mácsik J, Maurice C, Öhlander B (2015) Improving properties of sealing layers made of till by adding green liquor dregs to reduce oxidation of sulfidic mine waste. Geotechnical and Geological Engineering, 33(4), 1047-1054.

Manso JM, Ortega-López V, Polanco JA, Setién J (2013) The use of ladle furnace slag in soil stabilization. Construction and Building Materials, 40, 126-134.

Morin KA, Hutt NM, Coulter TS, Tekano WM (2003) Case study of non-mining prediction and control of acid rock drainage – the Vancouver Island Highway Project. Proceedings from the Sixth International Conference on Acid Rock Drainage, Cairns, Australia p 14-17.

Rose AW, Barnes, HL (2008). Alkaline addition problems at the Skytop Interstate-99 site, central Pennsylvania. Proceedings America Society of Mining and Reclamation, p 927-949.

Sarkkinen M, Kujala K, Kemppainen K, Gehör S. (2016) Effect of biomass fly ashes as road stabilisation binder. Road Materials and Pavement Design, 1-13.

Segui P, Aubert JE, Husson B, Measson M (2012) Characterization of wastepaper sludge ash for its valorisation as a component of hydraulic binders. Applied Clay Science, 57, 79-85.