DIM-ESEE Conference

Red mud as geotechnical composite

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Abstract

Red mud (RM), a by-product of the Bayer process, is produced in large quantities worldwide. It is mainly disposed of in settling ponds or through dry stacking, both of which pose environmental risks due to instability, alkaline emissions, and leaching of potentially toxic elements (PTEs). Several efforts have been made to utilize RM as an additive in soil remediation or in the production of construction materials. However, due to its fine-grained texture, poor mechanical properties, and tendency to leach harmful substances, RM alone is unsuitable for direct use in construction. Nevertheless, its mechanical properties and workability can be significantly improved by mixing it with hydraulic or pozzolanic binders, such as calcareous ashes.

In the present study the use of calcareous paper mill ash (PA) as an additive to RM, for production of geotechnical RM/PA composite was studied. The results demonstrate that mixing RM slurry with PA produces a soil-like geotechnical composite with suitable workability and mechanical properties for use in earthworks, comparable to natural soils. Moreover, the RM/PA composite exhibits lower leaching of PTEs, in comparison to the RM, improving the environmental acceptability of RM for its use in construction.

This study confirms that RM composites, with addition of alternative binders, can be a viable construction material, which not only meet the geotechnical performance requirements for earthworks but also comply with Slovenian environmental standards. This approach offers a dual benefit: it reduces the environmental and economical burden associated with RM disposal while simultaneously decreasing the demand for natural construction materials.

Keywords: red mud; recycling; geotechnical composites; environment

1. Introduction

Aluminium (Al) is one of the most produced metals in the world (**Svobodova-Sedlackova et al. 2024**). Its global consumption is expected to increase at a compound annual growth rate (CAGR) of 3.33 %, increasing from 101 million tonnes in 2024 to 119 million tonnes by 2030 (**URL 1**). Primary aluminium production is performed in two stages, via Bayer and Hall-Héroult processes. During the first stage, under Bayer process, alumina is extracted from the bauxite ore and RM is generated as a waste residue (**Svobodova-Sedlackova et al. 2024; URL 2**). The process is based on the digestion of the bauxite ore in a hot concentrated sodium hydroxide (NaOH) solution. The alumina-rich solution is then separated from the solid residue, which consists predominately of iron (Fe) and Al (hydr)oxides and is referred to as RM. The global production of RM amounts to more than 160 million tonnes per year (**Zalar Serjun et al., 2018; Pavšič et al., 2024; URL 2**).

Red mud has hazardous properties, which mostly result from its high pH (**Nayak et al., 2024**), as well as from the total content and leaching of potentially toxic elements (PTEs), and alkaline airborne dust emissions (**Jha et al., 2020; Samal, 2021**). Due to its colloidal structure, the containment and handling of RM pose a significant environmental problem (**Jha et al., 2020; Nayak et al., 2024; Pavšič et al., 2024**).

Therefore, potential applications for the use of RM need to be explored in order to minimise the associated negative environmental impacts (**Zalar Serjun et al., 2018; Jha et al., 2020**). However, the chemical and physical - mechanical properties of RM vary considerably and reflect the different sources - differences in the Bauxite composition and processing plant operation (**Jha et al., 2020; Oprčkal et al., 2020**). This means that from the scope of recycling each RM deposit must be considered case-by-case (**Oprčkal et al., 2020**). Various studies have shown that RM is a potential secondary source for the recovery of metals, such as iron (Fe), titanium (Ti), manganese (Mn), sodium (Na), potassium (K) and also various rare earth elements (REE) (**Samal, 2021**), but residual RM, after extraction of theses raw materials can also be regarded as a source of construction material (**Jha et al., 2020; Samal, 2021; Pavšič et al., 2024**). Due to its fine-grained nature and poor mechanical properties, as well as environmental concerns, RM cannot be used as a construction material by itself. The properties of RM residues can be significantly improved, when mixed with hydraulic or pozzolanic binders, such as calcareous ashes, to produce mechanically stable and environmentally acceptable composites for earthworks (**Jha et al., 2020; Pavšič et al., 2024**).

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Based on the results of laboratory tests, this study presents a useful utilisation of RM slurry by producing a geotechnical composite with addition of calcareous paper mill ash (PA), which is also an industrial residue, and offers dual benefits: It reduces the environmental impact associated with RM and PA disposal while reducing the demand for natural construction materials.

2. Materials and methods

Since PA and RM can be environmentally problematic (**Jha et al., 2020; Oprčkal et al., 2020**), the leaching of the PTEs, was investigated. The mineralogical composition of the PA and RM used in preparation of the RM/PA composite was also determined, since it defines the nature and behaviour of both materials in composite. Leaching of PTEs, mineralogical changes and basic geotechnical properties of the RM/PA composite were determined in order to evaluate the properties of the composite from the scope of potential use in earthworks and to investigate the influence of the PA addition on environmental and mechanical properties of the RM/PA composite in comparison to the RM itself.

2.1. Materials

In this study the sample of red mud (RM) from the old pond in Podgorica (Montenegro) after gravitational and magnetic large-scale separation of raw materials (metals, REE) was used. Residues were first left undisturbed for a longer period of time so that particles were able to settle. This was followed by decantation of the supernatant liquid. The residues used for investigations were in the form of slurry with a moisture content of 139 wt.%, which represents a 42 wt.% of solids in the slurry.

Fly ash from paper mill (PA) is a recycled material with hydraulic and pozzolanic properties that can be used as an alternative binder for soil remediation, solidification and stabilization. It is a highly alkaline material that contains a (latent) hydraulically active phase. The composition and properties of PA are highly dependent on the type of fuel burned, the combustion technology used, and the combustion conditions, which could affect the properties of the geotechnical composite material produced from it. In this study, a PA from the paper mill Lenzing AG, produced from burning of deinking sludge and waste cellulose fibre from recycling of paper was used.

2.2. Methods

A geotechnical composite, designated as RM/PA was prepared by mixing RM slurry with PA. The amount of PA required to produce the geotechnical composite was determined on basis of laboratory trials. Quantity of added PA with which the appropriate consistency of the RM/PA for the compaction was achieved was selected. The RM/PA mixture was prepared by homogenizing RM and a selected amount of PA, with a hand-held screw-mixer. For the preparation of RM/PA geotechnical composite, 0.75 kg of PA was added to each 1 kg of RM slurry.

Prior to the RM/PA specimen preparation, the mellowing period of 24 hours was required to allow the chemical reactions to take place and to achieve suitable consistency - workability of the mixture. During the mellowing period, the mix was stored in an airtight plastic container at ambient temperature. After the mellowing period, the mixture was homogenized again and the test specimens were prepared by compaction in the molds according to **SIST EN 13286-50 (2005)** using the standard Proctor compaction effort (**SIST EN 13286-2, 2013**). The specimens were removed from the mold by a press and then cured at 22 °C and 98 % humidity for the selected period of time. The prepared specimens were used to determine the geotechnical, mineralogical and chemical properties of the investigated geotechnical composite at selected curing times.

Bulk mineralogical composition of the PA, RM and RM/PA composite was determined by using X-Ray Powder Diffraction (XRD) using an EMPERYAN (PANalytical) X-ray diffractometer equipped with Cu Kα radiation. Data were collected at 45 kV and 40 mA in the range 5–70° 2θ, with a 0.013° 2θ step size and measuring time per step of 148.9 s. Interpretation of the results was carried out by means of Highscore v.4.x LTU software, involving PDF 4 database. Prior to the XRD analysis, the subsamples were dried up to a constant mass and pulverised in an agate mortar.

The leaching tests on PA, RM and RM/PA composite after 7 days of curing, were performed according to **SIST EN 1744-3 (2002)**, at the liquid to solid ratio of 10:1. The preparation of the eluates was carried out in high density polyethylene (HDPE) beaker.

Concentrations of chromium, cobalt, nickel, copper, zinc, arsenic, selenium, molybdenum, cadmium, antimony, barium, mercury and lead in eluates, were measured by inductively coupled plasma mass spectrometry (ICP-MS), according to **SIST EN ISO 17294-2 (2017)**. The content of chloride and sulphate was defined by spectrophotometry according to **ISO 15923-1 (2013)**, while the fluoride content was defined according to the 4500-FD – colorimetric SPADNS method (**SMWW, 2017**).

Limiting values of the PTEs in RM/PA eluates were taken from the Slovenian national legislation according to the Waste decree – Annex 5: Permissible levels for pollutants in their leachates for processed substances or objects that will be used in the external environment and exposed to atmospheric influences and have the property of leaching with permeability $\leq 10^{-9}$ m/s (**URL 3**).

Geotechnical properties of RM/PA composite, such as the unconfined compressive strength (Rc) (**SIST EN 13286-41, 2022**), the consolidation properties (**SIST EN ISO 17892-5, 2017**), permeability (**SIST EN ISO 17892-11, 2019**), and shear strength (**SIST EN ISO 17892-10:2019**) were determined after the selected time of curing (2 days, 7 days and 14 days for Rc, and 2 days for the others), while the moisture content (**SIST EN ISO 17892-1, 2015**) was determined at preparation.

3. Results

For the preparation of the RM/PA a RM from Podgorica pond after extraction of raw materials, with moisture content of 139 % wt. (42 % wt. solids) and PA from paper mill Lenzing AG were used. The mineralogical compositions of RM, PA and RM/PA composite after 7 days of curing are presented in **Table 1** and in **Figures 1 - 3**, while eluates quality for both materials, and the composite after 7 days of curing, alongside the legislative limits (**URL 3**) are presented in **Table 2**.

Table 1. Mineralogical composition of RM and PA

Mineral phases		
RM	PA	RM/PA
Hematite (Fe_2O_3)	Anhydrite ($CaSO_4$)	Gypsum ($CaSO_4x2H_2O$)
Gibbsite ($Al(OH)_3$)	Lime (CaO)	Hematite (Fe_2O_3)
Cancrinite $(Na,Ca)_8(Al_6Si_6O_{24})(CO_3,SO_4)_2 x2H_2O)$	Quartz (SiO_2)	Calcite ($CaCO_3$)
Calcite ($CaCO_3$)	Calcite ($CaCO_3$)	Quartz (SiO_2)
Boehmite ($AlO(OH)$)	Magnetite (Fe_3O_4)	Gibbsite ($Al(OH)_3$)
Aluminium fluoride (AlF_3)	Portlandite ($Ca(OH)_2$)	Cancrinite $(Na,Ca)_8(Al_6Si_6O_{24})(CO_3,SO_4)_2x2H_2O)$
	Periclase (MgO)	Portlandite $Ca(OH)_2$)
	Gehlenite ($Ca_2Al(AlSiO_7)$)	Gehlenite $Ca_2Al(AlSiO_7)$)
		Ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12} x26H_2O$)

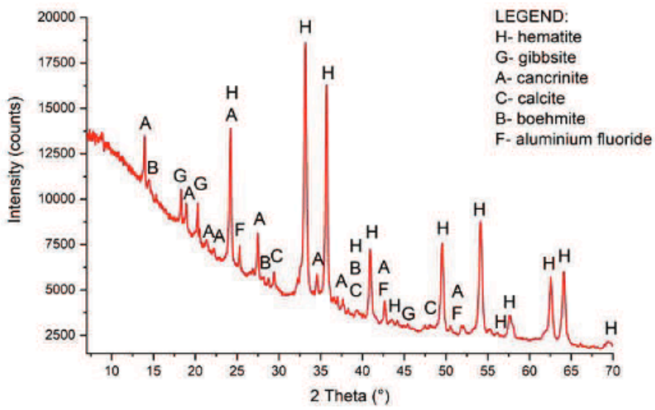

Figure 1. XRD spectrum of RM

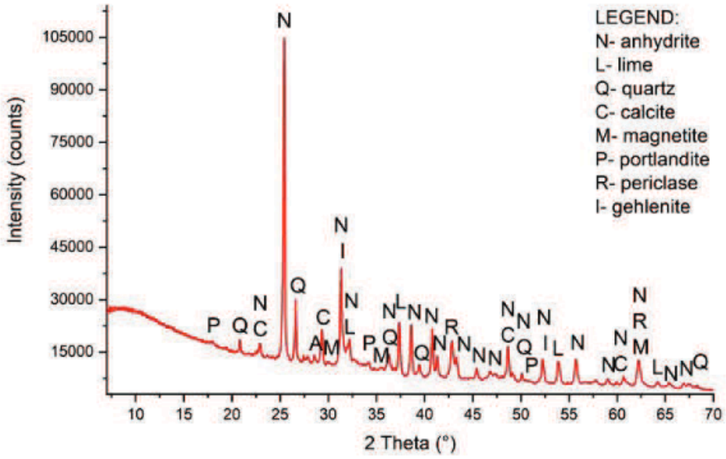

Figure 2. XRD spectrum of PA

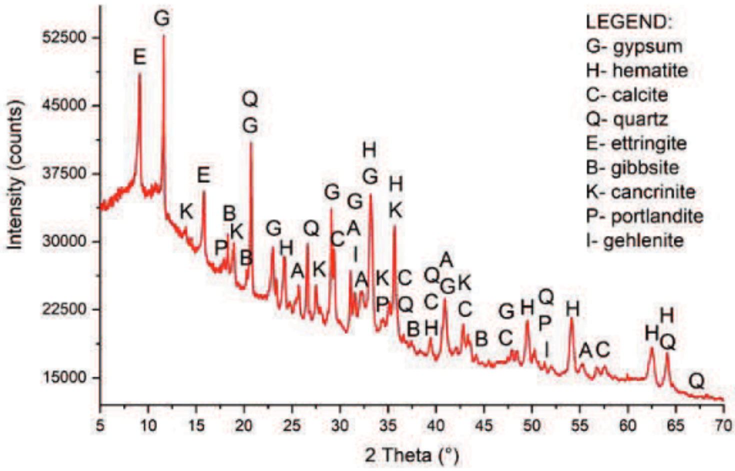

Figure 3. XRD spectrum of RM/PA

Table 2. Chemical analyses of eluates from RM, PA and RM/PA

Parameters	Concentration (mg/kg $_{d.m.}$)			
	RM	PA	RM/PA	Limit values (**URL3**)
Chromium (Cr)	0.03	0.03	0.02	0.6
Cobalt (Co)	0.003	0.006	0.002	0.05
Nickel (Ni)	0.02	0.01	<0.01	0.5
Copper (Cu)	0.03	0.113	0.106	2
Zinc (Zn)	<0.002	22.2	<0.002	3.5
Arsenic (As)	0.87	0.01	0.01	0.4
Selenium (Se)	0.02	<0.002	<0.002	1
Molybdenum (Mo)	0.02	1.14	0.09	1
Cadmium (Cd)	<0.001	0.002	<0.002	0.04
Antimony (Sb)	0.004	<0.002	0.014	0.5
Barium (Ba)	0.01	1.83	0.52	20
Mercury (Hg)	<0.001	0.001	0.001	0.01
Lead (Pb)	0.009	0.115	0.003	0.6
Fluoride (F^-)	16	520	<5	10
Chloride (Cl^-)	62	3720	210	1000
Sulphate (SO_4^{2-})	131	22860	3480	10000

The geotechnical properties of the compacted RM/PA composite are presented in **Table 3**.

Table 3. Geotechnical properties of RM/PA composite

Parameter	Method	Result
Moisture content	SIST EN ISO 17892-1:2015	39.7 %
Dry density of compacted sample ρ_d	SIST EN 13286-2:2010/AC:2013	1.14 Mg/m^3
Unconfined compressive strength, tested after curing time:	SIST EN 13286-41:2022	
- 2 days		144 kPa
- 7 days		3548 kPa
- 14 days		4939 kPa
Consolidation properties – Oedometer test Vertical stress:	SIST EN ISO 17892-5:2017	
- 50 kPa		3560 kPa
- 100 kPa		79400 kPa
Permeability k_{20} at 200 kPa	SIST EN ISO 17892-11:2019	5.5×10^{-11} m/s
Shear strength	SIST EN ISO 17892-10:2019	
- Friction angle φ'		51.5 °
- Cohesion c'		0.0 kPa

4. Discussion

The major mineral phases of the RM used are hematite, gibbsite and cancrinite, while calcite, boehmite and aluminium fluoride are in the minority. The leaching tests have shown that the concentrations of As and F^- in the eluate of RM exceed the limits specified in the Slovenian environmental legislation (**URL 3**). The mineralogical analysis of the PA used in preparation of the composite shows that, it contains, among other minerals, anhydrite, which could affect the mechanical stability of the RM/PA composite due to the volumetric changes during the transformation into gypsum and also causes high concentrations of sulphate in the PA eluate. Lime (CaO) and periclase (MgO) are present in the PA as active phases that can form hydration products. The leaching test shows an access concentrations of Zn, Mo, F^-, Cl^- and SO_4^{2-} (**URL 3**) in the PA eluate.

The mineralogical composition of the RM/PA composite shows that a new mineral phase - ettringite - was formed during the curing of the investigated geotechnical composite, which is associated with the immobilisation of various PTEs (**Kumarathasan et al., 1997**). The immobilization effect, which could be at least partially contributed to ettringite formation, is confirmed by the RM/PA eluate, in which the concentrations of As and F^- from RM and of Mo, Zn, F^-, Cl^- and SO_4^{2-} from PA were reduced below the Slovenian legislative limits (**URL 3**), which is also partly due to the formation of insoluble species, thus ensuring the environmental acceptability of RM/PA. However, despite the favourable results of RM/PA eluate quality, the long-term environmental acceptability of RM/PA could be affected by exposure to various environmental factors. Although the possibility of environmental degradation of the RM/PA composite is very low due to its high alkalinity, assumed high buffering capacity and low permeability, which reduces the potential migration of PTEs, its long-term and pH-dependent behaviour should be further investigated.

The geotechnical composite RM/PA exhibits high shear strength and low compressibility and permeability. The uniaxial compressive strength increases with the age. After 7 days of curing, the increase in strength slows down, indicating that most of the hydration process has been completed. The early Rc value of RM/PA is relatively low, but after 7 days of curing the minimum Rc value of 0.4 MPa is exceeded (**SCS, 1989**). From the point of view of evaluated geotechnical properties, the investigated composite material shows even better properties than most natural soils, which confirms its applicability as a construction material.

5. Conclusions

In this study, the utilization of RM with addition of PA as a geotechnical composite material was investigated. Since the RM residue is in the form of slurry, it cannot be used as a geotechnical material by itself, but must be treated with binders to obtain a geotechnical composite material with appropriate geotechnical properties. An alternative recycled material, PA, containing (latent) hydraulically active phases was selected as an additive to the RM slurry in a geotechnical composite RM/PA. A material with the desired properties for use in earthworks was obtained, with even better geotechnical properties than most natural soils used in earthworks. During the production of the samples, a mellowing period was needed to gain proper workability of the investigated composite. The composition of the binder also affects the leaching of potentially hazardous substances and thus the environmental acceptability of the composites. Therefore, the alternative binder should be carefully selected according to the properties that must be suitable for the composite production and should be carefully investigated in advance.

The results of this study show that RM after extraction of raw materials (metals, REE) could be used as an alternative construction material in earthworks, if it is treated with appropriate additives or binders, such as paper mill ash, to produce geotechnical composites. Such a composite could be manufactured and marketed as a construction product after obtaining technical approval at European or national level, depending on its intended use and properties.

Nevertheless, red mud composites need to be further investigated, especially with regard to the evaluation of long-term stability under changing climatic conditions.

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Acknowledgment

The authors acknowledge the Slovenian Research and Innovation Agency core funding P2-0273 and project J1 - 4413.

Funding

This research was funded by EIT RawMaterials project, RIS-RESTORE, project no. 19269, co-funded by the European Union.

Author's contribution

Primož Pavšič (PhD): investigation, writing – review & editing. **Marija Đurić** (PhD): investigation, writing – review & editing. **Mateja Košir** (PhD): conceptualization, funding acquisition, investigation, writing – review & editing. **Primož Oprčkal** (PhD): conceptualization, funding acquisition, investigation, writing – review & editing. **Vesna Zalar Serjun** (PhD): conceptualization, investigation, writing – review & editing.

All authors have read and agreed to the published version of the manuscript.