



Physical properties of a soil substitute derived from an aluminum recycling by-product

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Abstract

Disposal of highly saline industrial by-products in landfills is not permitted in member states of the European Union, such as Germany. Large amounts of such by-products thus have to be disposed of in alternative ways. In many countries bare potash mining residue mounds, consisting almost entirely of rock salt (NaCl), pose environmental problems. Covering such mounds with soil or soil-like material could help to reduce the yearly amount of briny runoff. A fine-granular saline aluminum recycling by-product (ALRP) has been proposed as a soil substitute to cover rock salt residue mounds. Use of this by-product as a combined soil substitute and surface barrier is not considered to be a landfill disposal, but as a beneficial by-product reuse. To judge the feasibility of ALRP for this purpose, its properties must be known. In this study physical characteristics of an industrially produced ALRP, mixed with the flue gas desulfurization by-product (FGDP) of a coal combustion power plant, were determined. It was found that the texture of both ALRP and ALRP–FGDP mix was silt loam. Bulk densities of ALRP and ALRP–FGDP were 0.93 and 0.88 Mg m⁻³ and the corresponding salt contents were 50.0 and 35.5%, respectively. The erodibility factor *K* of pure ALRP was estimated as 0.65 Mg h ha⁻¹ N⁻¹. Because of the stabilizing effect of FGDP, this factor was reduced considerably in ALRP–FGDP. The water-holding capacity of unwashed ALRP was 44.5% and of washed ALRP–FGDP 61.8%. In view of its physical properties, ALRP–FGDP seems to be suitable as an evaporation enhancing, runoff reducing cover material for potash mine residue mounds, even on steep slopes. Use of ALRP, mixed with FGDP, as a soil substitute in a surface barrier, thus seems to be environmentally meaningful. However, the high salt content initially prevents plant growth. With time, after the salt has been leached, the material seems able to support plant

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growth, which would further reduce runoff. The physical and hydraulic parameters determined in this study may serve future users of similar by-products.

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1. Introduction

Potash mining, providing agriculture with an essential plant nutrient, has a long tradition in Germany. After the theory of mineral nutrition of plants had widely gained acceptance around 1850 [1], mining operations started in 1861 at Stassfurt in East Germany. Since then, potash mining has been practiced at many locations in Germany, for example, around Hannover in the northern part of the country. With time, large residue mounds have been piled up. In this study we report on the potash mine Sigmundshall, west of Hannover. At the site of this mine, operated by the K+S Company (Kali und Salz GmbH), a mound of 120 m height, containing approximately 13.4 million m³ of rock salt, has been piled up. This mound, with its steep ($\approx 35^\circ$) and crusted slopes, is completely bare, because it consists over 95% of NaCl.

Under the humid climate of Northern Germany, the mound produces yearly around 150 000 m³ of briny discharge, which is conveyed into the nearby Leine river. The yearly amount of NaCl carried into this river is estimated as 54 Gg. Other potash mines around Hannover cause similar environmental problems. Because the mound's surface hardly stores any moisture during rainfall, evaporation from the mound is low. It has therefore been suggested to cover the mound with soil or soil-like material in order to store temporarily near-surface soil moisture during rainfall events. Subsequently, this soil water can (at least partly) evaporate, thus decreasing the amount of briny runoff. Because soil is a valuable, but increasingly scarce resource, a fine-granular dark-gray and highly saline (NaCl) aluminum recycling by-product (ALRP), possibly mixed with other industrial by-products, has been proposed as a soil substitute.

Aluminum for industrial use is provided by two different suppliers: Primary Al producers and secondary, recycling Al smelters. In most applications, recycled Al performs equally well as primary Al. Because secondary Al is produced with as much as 90% energy, capital, and labor savings, in comparison with primary production [2], an increase in Al recycling has been observed worldwide. In the US, for example, the production of recycled Al rose from 850 Gg in 1986 to 1500 Gg in 1998 [2]. Other countries with a high secondary Al production are Germany, Italy, Japan, UK, France, and The Netherlands. In Germany, 46% of the total 709 Gg of Al produced in 1996, was recovered from scrap [3].

During the recycling process, Al scrap commonly is melted in reverberatory or rotary furnaces. At the charging well of the furnace, the scrap is lowered through a layer of molten salt, called flux. The salt flux layer in the furnace serves multiple purposes. It enhances heat transfer to the scrap, strips oxides and other impurities such as carbides, nitrides, and sulphides from the melting metal, and keeps inclusions in suspension. By providing a physical barrier between the air and Al, the salt flux minimizes Al oxidation. Fluxes of

various chemical compositions have been developed. Besides reducing losses, it is believed that selected fluxes are effective in refining the metal from different types of scrap. Most commonly, the salt flux is a mixture of NaCl (about 70 wt.%) and KCl (about 30 wt.%). Salt flux also can contain small amounts of CaF_2 [4].

The amount of salt used depends on the amount of impurities contained in the scrap. For each kilogram of impurities, approximately 1 kg of flux is used. When liquified the metal is tapped from the furnace, and the top layer (which consists of solid Al_2O_3 , spent flux, entrapped metal, and other contaminants) must be skimmed. This top layer of dark-gray material is generally called black dross. Typically, black dross contains metallic Al (10–20 wt.%), a salt flux mixture (40–55 wt.%), and 20–25 wt.% of Al_2O_3 [5]. Depending on the market price of Al and the cost of landfill, black dross can be processed further to recover as much as 20% of the remaining Al by means of salt bath rotary furnaces or hammer mills, which separate Al physically from the dross. When the recovery of Al from dross is not economically feasible, and if environmental regulations permit dumping, dross is usually disposed of in conventional landfills.

During black dross processing, low-grade scrap and black dross are melted in rotary furnaces. Because of the high content of impurities, considerably more flux is used during black dross processing as compared with recovering Al from high-grade scrap. The residues that are generated during black dross processing are referred to as salt cake. The composition of salt cake depends on that of the black dross; often it contains 3–5 wt.% Al, 15–30 wt.% Al_2O_3 , 30–55 wt.% NaCl, and 15–30 wt.% KCl. The amount of black dross and salt cake, produced per 1000 kg of recovered Al, ranges from 400 to 700 kg. Worldwide, considerable amounts of black dross and salt cake are produced yearly; for example, 510 Gg of salt cake were generated in the US in 1993 [6]. If permitted, also salt cake is disposed of in landfills.

Because of environmental concerns, however, the disposal of dross and salt cake in conventional landfills in some European countries is not allowed [7]. In member states of the European Union, such as Germany, the disposal of black dross and salt cake in landfills is not permitted, because these wastes do not fulfill the acceptance criteria determined in accordance with Annex II of the Council Directive 1999/31/EC on the landfill of wastes [8]. Therefore, recycling systems for those wastes, as well as alternative means of disposal have to be developed. The present study reports on such an alternative means of disposal.

In 1996, a black dross and salt cake recycling plant opened at the Sigmundshall potash mine. Besides potash, the mine produces salt flux for the secondary Al industry. This industry returns its black dross and salt cake production to the Sigmundshall recycling plant. The plant has a yearly processing capacity of 90 Gg black dross and salt cake. Annually, about 3.6 Gg of metallic Al is separated from raw material in a dry processing stage that involves both crushing and screening. In a subsequent wet processing stage, KCl is leached at a temperature of 140 °C and 8.7 Gg of KCl are recrystallized by cooling the hot brine in a vacuum atmosphere to 35 °C. The crystallized potash is dried and sold as a fertilizer containing an equivalent of about 50% of K_2O . Sodium chloride remains in the solution, which is reused for the leaching process. In the present paper, we refer to the solid granular residue of recycled black dross and salt cake as aluminum recycling by-product, or ALRP. Mainly because of the high soluble salt content (NaCl), also the disposal of ALRP in landfills

Table 1
Principal components and trace elements in the ALRP used in the present study^a

Element	Amount (wt.%)	Element	Amount (wt.%)	Element	Amount (mg kg ⁻¹)
Al	18.78	Na	13.65	Cu	2314
Si	2.40	Cl	18.19	Cr	417
Ca	1.58	P	0.02	Ni	169
Fe	0.77	S	0.18	Pb	116
K	2.53	F	0.33	Zn	708
Mn	3.66	Ti	0.29	As	4

^a Ignition loss was 13.79%; Mg was not determined.

is not permitted [8]. Main components of ALRP are Al₂O₃ (27–33 wt.%), SiO₂ (4–10 wt.%), and soluble NaCl (45–55 wt.%). Magnesium aluminate (MgAl₂O₄), Fe₂O₃, CaO, CaF₂, and KCl (1–3 wt.%) also may occur in ALRP. By centrifugation, the material is dewatered to a water content of 16–18 wt.%. Subsequently, it is used as the main component in the cover material for the nearby rock salt residue mound. Use of ALRP as a soil substitute in a surface barrier over a mining residue mound of mainly rock salt (NaCl) is not considered as a landfill disposal, but as an environmentally meaningful reuse of an industrial by-product. For details on the salt leaching of the cover material under field conditions, the reader is referred to Hermsmeyer et al. [9,10].

Table 1, prepared from X-ray fluorescence analysis data, shows the chemical composition of air-dry ALRP. Values indicate that Al and Si are main constituents of ALRP, but also that the amounts of Na and Cl are high. The table indicates further that ALRP contains a number of trace elements, some of them (e.g. Cu and Cr) occurring in concentrations that trigger environmental concerns when ALRP is proposed as a soil substitute.

As mentioned above, the dross processing plant at Sigmundshall has been producing ALRP since 1996. On the north slope of the mine's NaCl residue mound, an area of about 2 ha was assigned for experiments with ALRP as cover material. Initially, pure ALRP was transported with a conveyor belt up the mound and placed on the test site below. Later, after it was observed that pure ALRP on the steep slopes was too susceptible to slumping and erosion by water, ALRP was mixed with the flue gas desulfurization by-product (FGDP) of a coal combustion power plant. Flue gas desulfurization at coal combustion power plants involves the removal of SO_x from the smokestack, usually by adding a limestone slurry to the flue gas stream. As a result, FGDP is a combination of fly ash, Ca–S salts (CaS and/or CaSO₄), and CaCO₃. Actual composition of FGDP may vary considerably, depending on the parent coal, the conditions of combustion, and the type of emission control devices, including the desulfurization system used [11,12]. In construction engineering it is well known that FGDP has stabilizing, cementing properties. For example, the FGDP described by Laperche and Traina [13] was used recently for construction of embankments and livestock waste lagoons.

Primary objective of the present study was to determine the main physical and hydraulic properties of ALRP, FGDP, and ALRP–FGDP and to assess the geotechnical feasibility of ALRP and ALRP–FGDP as cover materials for the bare and steep Sigmundshall potash mine residue mound. Of particular interest were the water-holding capacity of the tested materials and the susceptibility to slumping and water-induced erosion.

2. Materials and methods

The Sigmundshall black dross and salt cake recycling plant and a coal combustion power plant from Hannover provided the original materials for the analysis. These included, besides ALRP and FGDP, a volumetric mixture of seven parts ALRP and three parts FGDP, called ALRP–FGDP. Samples of several kg were transported to the laboratory in sealed containers and were stored at 10 °C. Properties determined included moisture and salt content, particle density (PD) and bulk density (BD), total porosity, particle size distribution, saturated conductivity for water and salt solution, moisture retention at various suctions, consistency limits, and erodibility. Unless specified otherwise, standard methods of analysis [14] were used.

Some analyses involved saturation or intensive mixing of samples with water. With high salt-content materials, this led to a partial sample loss, because of dissolving and leaching. Therefore, some methods were only applied to salt-free (washed) samples. To produce such samples, the material was washed with deionized water in an overhead shaker at room temperature. Solution and solids were separated in a centrifuge, and electrical conductivity (EC) was measured in the solution. The solution was replaced by fresh water, and washing was repeated until EC was lower than 0.6 mS cm⁻¹ (tap water). Besides water, brine from the first washing cycle, in equilibrium with the corresponding material, was used for measurements of saturated hydraulic conductivity.

2.1. Moisture and salt content

Moisture content was determined as follows. Samples were weighed in metal cups and oven-dried at 105 °C to constant weight [14]. Moisture content was calculated as a percentage of total sample weight and is reported on a dry solid basis including salts.

Stainless steel rings of 56 mm inside diameter and 40 mm height were used to determine volumetric moisture content, BD, moisture retention, and saturated hydraulic conductivity. A thin dish-washing cloth was placed at one end of each ring and held in place with a rubber band. Each ring was filled one-third and compacted (slightly) with a steel cylinder; this procedure was repeated until the rings were completely filled. Samples were weighed and oven-dried at 105 °C. Moisture content was calculated as a percentage of sample volume.

The salt content of the test materials was calculated from the EC of aqueous solutions. Small samples of oven-dry material (≈ 10 g) were mixed with deionized water and shaken thoroughly. Suspensions were left standing for 0.5 h in order to let undissolved particles settle. Subsequently, EC was measured in the solution over the solids. Assuming that a rise in EC of 1 mS cm⁻¹ corresponded to a rise in salt concentration of 0.64 g l⁻¹ [15], we calculated the salt content S (in wt.%) as

$$S = 0.064 \text{ EC} \frac{w}{m} \quad (1)$$

where w (in g) is the amount of water added, and m (also in g) is the weight of the oven-dry sample. Initial measurements showed that the ratio of oven-dry material to water had to be at least 1:7 in order to dissolve all solid-soluble components. In view of this experience, a ratio of 1:10 was chosen for all determinations of salt content.

2.2. Particle density (PD), bulk density, and total porosity

PD of washed samples was determined with the pycnometer method, described by Blake and Hartge [16]. PD of samples including salt was calculated from PD of both salt and nonsalt components. Percentages of nonsalt and salt components were used as weighting factors. Because the salts in ALRP consisted primarily of NaCl, the PD of the salt was assumed to be 2.165 Mg m^{-3} , which is the density of NaCl [17]. Following Blake and Hartge [18], packed ring samples as described earlier were oven-dried, weighed, and BD was calculated as weight divided by ring volume. Total porosity was calculated from PD and BD [19] as

$$\text{Porosity (vol.\%)} = \left(\frac{1 - \text{BD}}{\text{PD}} \right) 100 \quad (2)$$

2.3. Particle size analysis

Particle size analysis was carried out according to Gee and Bauder [20]. Samples were mechanically disaggregated to pass a 2 mm sieve. Weighed samples ($\approx 10 \text{ g}$) were sieved into the following classes: 0.63–2, 0.2–0.63, and 0.063–0.2 mm. Stokes' sedimentation procedure was used to partition the fraction $< 0.063 \text{ mm}$ into the following classes: 0.02–0.063, 0.0063–0.02, 0.002–0.0063, and $< 0.002 \text{ mm}$ (clay). Fractions were oven-dried and weighed, and the difference between initial sample weight and the sum of all fractions was assumed to be the initial salt content of the sample.

Particle size distribution data for the FGDP were provided by the GfR Company (Gesellschaft für die Aufbereitung und Verwertung von Reststoffen, mbH), Würzburg, Germany. A laser-light scattering method as described by Allen [21] or Cooper et al. [22] was applied, using a Malver Diffraction Particle Sizer, Type Mastersize S.¹ No pretreatment of samples was conducted.

Particle-size distribution for salt-free ALRP–FGDP was computed, using results from pure ALRP and pure FGDP, with the fractions of the mixing ratio as weighting factors. Fractions were 7:3 on a volumetric basis for saline ALRP and FGDP, which corresponds to 5.2:3 for the washed mix, when an initial salt content of ALRP of 50 wt.% is assumed.

2.4. Saturated hydraulic conductivity and intrinsic permeability

Saturated hydraulic conductivity, K_s , was measured using the constant-head method [23]. Water was used as flow medium for both washed and unwashed samples. Additionally, salt solutions in equilibrium concentration with the investigated material were used to determine K_s of unwashed samples in order to reduce leaching losses during the measurement. Each K_s measurement was repeated six times. In addition to K_s , the intrinsic permeability $k_{w(\text{or s})}$ was determined as

$$k_{w(\text{or s})} = K_{S_{w(\text{or s})}} \frac{\eta_{w(\text{or s})}}{\rho_{w(\text{or s})} g} \quad (3)$$

¹ Use of brand names is for information only and does not imply endorsement of products by the authors of this article.

where η is the dynamic viscosity, ρ the fluid density, and g the acceleration due to gravity. The subscript w (or s) in Eq. (3) denotes that either water or a salt solution was used as fluid. The density $\rho_{w(\text{or } s)}$ of the fluids was measured with pycnometer flasks and the viscosity $\eta_{w(\text{or } s)}$ with a capillary flow viscosimeter, submerged in a water tank, kept at a constant temperature of 20 °C. Intrinsic permeability is a property of the medium, supposedly independent of the fluid, and has the dimension of length squared (L^2). For comparison with older soil literature, we express permeability in units μ^2 ($= 10^{-8} \text{ cm}^2$).

2.5. Moisture retention

Moisture retention was measured with standard procedures [14] on packed core samples of unwashed and washed ALRP, washed FGDP, and washed ALRP–FGDP. Samples were carefully saturated from the bottom with de-aerated tap water, placed on a porous ceramic plate and dewatered with a hanging water column in steps of 0.1, 0.3, 0.6, and 1.0 m, and a kaolin box with a hanging Hg column for 3.0 m. A pressure plate apparatus was used at 158 m (permanent wilting point) to drain washed samples of 1 cm height. Water-holding capacity was defined as the water content at 1.0 m of suction.

Because of salt loss during saturation and dewatering, unwashed ALRP samples settled during the measurement, thus influencing the result. Settling was recorded, and changes in sample volume and weight, salt content, BD, and volumetric water content were estimated after each dewatering step, using average PD values for soluble and nonsoluble sample components, and assuming that loss of sample volume was equal to volumetric salt loss. The RETC code [24] was used to estimate residual water content θ_r , saturated water content θ_s , and empirical van Genuchten parameters α , n , and m . The inverse of the empirical parameter α (L^{-1}) is often referred to as the air entry value or bubbling pressure, i.e. the suction at which the material begins to release significant amounts of water, or the suction at which the water retention curve bends downward. To impose a restriction on the permissible values of the empirical constants m and n , the relation $m = 1 - (1/n)$ was selected. Parameters α , n , and m were used to fit retention curves of the form [25]

$$S_e = \frac{1}{[1 + (\alpha h)^n]^m} \quad (4)$$

to the measured data, where S_e is the effective degree of saturation or reduced water content ($0 \leq S_e \leq 1$), and h is the suction.

2.6. Consistency, erodibility, and erosion

The consistency or Atterberg limits (liquid and plastic limit), were determined by standard procedures [26]. The plasticity index was calculated as the difference between water content at the liquid and the plastic limit [27].

The susceptibility of ALRP to water-induced erosion was assessed with use of the Revised Universal Soil Loss Equation (RUSLE). This equation, as well as the original Universal Soil Loss Equation (USLE) can be given [28,29] as

$$A = R K L S C P \quad (5)$$

where A is the average annual soil loss (Mg ha^{-1}), R the rainfall erosivity index ($\text{kJ mm m}^{-2} \text{h}^{-1}$), K the soil-erodibility factor ($\text{Mg h ha}^{-1} \text{N}^{-1}$), LS is the topographic factor, C the cropping-management factor, and P the conservation practice factor.

The factors needed for computational work with Eq. (5) were estimated following [30]. The erosivity index R was calculated from the mean annual rainfall at the site of the mound. The erodibility factor K of ALRP was estimated from its texture, organic matter content, aggregate size, and permeability class with use of the relation [30]

$$K = 2.77 \times 10^{-6} M^{1.14} (12 - \text{OM}) + 0.043(I - 2) + 0.033(4 - J) \quad (6)$$

where $M = (\% \text{silt} + \% \text{very fine sand})(\% \text{silt} + \% \text{sand})$, OM is the percentage of organic matter, I the class of aggregate size, and J the permeability class.

The value for M in Eq. (6) was calculated from the data of the particle size analysis for ALRP. For details we refer to [30]. Because of the high temperature under which ALRP is produced, we assumed that ALRP did not contain organic matter ($\text{OM} = 0.0$). Before crushing and sieving the material for particle size analysis, aggregates of about 2–10 mm in diameter were observed. According to [30], this corresponds to a class of aggregate size denoted as $I = 3$. In view of the measurements of the hydraulic conductivity, the permeability class was set = 2 ($J = 2$). Slope length and steepness factors L and S of the mound were derived from an analysis of the geometry. The geometry of the mound was determined with the use of stereoscopic aerial photographs, a Digital Terrain Model of $5 \text{ m} \times 5 \text{ m}$ cell resolution [31], and a cell-based geographic information system (GIS) integrated modeling technique. The continuous function for slope steepness influence on soil loss [32] was used to calculate S . Cropping management factor C and conservation practice factor P were assumed to be unity, because initially the ALRP-covered test field was bare and no conservation measures to reduce erosion were taken after the cover material was deposited on the slopes of the mound. With the RUSLE factors thus estimated, the mean annual material loss due to water erosion was calculated. Actual losses were not measured, but assessed visually after major rainfall events.

3. Results and discussion

3.1. Moisture and salt content

During early stages of ALRP production at Sigmundshall, the material was dewatered under pressure. An average moisture content of 35% was achieved, which turned out to be too high for the material to be transported on the upslope conveyor belt. A moisture content of the ALRP–FGDP mixture of 12–18% was yielding a product of optimal coherence for transport and distribution. Because the available FGDP had a moisture content of <3% and the mixing ratio of ALRP and FGDP was set to 7:3, the moisture content of ALRP had to be <25%. Since 1998, a decanting centrifuge has been operated to dewater ALRP to 18% before the materials are mixed. An average moisture content of 13% was determined in the material coming from the mixer, which is at the lower end of the optimum range of coherence. A sprinkler system has been installed to moisten the mixed material when dust is occurring.

Table 2
Salt content, PD, BD, and total porosity of unwashed and washed ALRP, FGDP, and ALRP–FGDP ($N = 3$)^a

Material	Salt content (%)		PD (Mg m^{-3})		BD (Mg m^{-3})		Total porosity (%)	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Unwashed								
ALRP	50.0	45.0–55.0	2.77	2.63–2.94	0.93	0.88–0.99	66.4	62.4–69.9
FGDP	1.65	1.1–2.3	2.31	2.27–2.35	0.68	0.67–0.70	70.6	69.2–71.5
ALRP–FGDP	35.5	32.0–39.0	2.46	2.31–2.67	0.91	0.66–1.10	63.0	52.4–75.3
Washed								
ALRP	<0.05	–	3.07	3.05–3.08	0.93	0.92–0.93	69.8	69.5–70.1
FGDP	<0.05	–	2.31	2.27–2.35	0.56	0.55–0.57	75.7	74.9–76.6
ALRP–FGDP	<0.05	–	2.67	2.67–2.68	0.88	0.87–0.88	67.2	67.0–67.5

^a The number of replicate measurements N in each case was three.

Salt content of ALRP–FGDP is a key parameter when the material is to be used as a soil substitute on which eventually plants have to grow. Average initial salt content was found to be 50% in unwashed ALRP, 1.65% in unwashed FGDP, and 35.5% in the unwashed mixture (Table 2). Salt contents calculated from EC measurements using Eq. (1) and loss of sample mass during particle size analysis varied by less than 4%. No controlled laboratory experiments with either ALRP or ALRP–FGDP mix were conducted to study the leaching dynamics of both materials. However, hydrological estimates can be made to predict the time required to establish vegetation.

At 20 °C, 357 g of NaCl is soluble in 1 l of water [17]. Average annual rainfall at the site is 640 mm and average annual potential evaporation is 550 mm [33]. If, for simplicity, potential evaporation is assumed equal to actual evaporation, average seepage is 90 mm per year. Since the BD of the mix is about 1 Mg m^{-3} (see Section 3.2), a layer of 0.5 m thickness and unit cross area (1 m^2) of ALRP–FGDP contains 177.5 kg (i.e. 35.5%) of easily soluble components, predominantly NaCl. If seepage is salt-saturated until all salts are leached, 483 mm of seepage (or 5.4 years) are needed to remove salts from a 0.5 m thick layer to a plant tolerable level, if we assume piston flow to occur. Our simplified discussion of desalinization neglects redistribution and reprecipitation of salts as a result of evaporation. In a lysimeter study of water and salt dynamics in ALRP and ALRP–FGDP we have observed desalinization of both materials over 3 years under humid field conditions. In the lysimeter study it was further observed that a grass cover was able to develop 28 months after the start of the experiment. Results of the lysimeter experiment are published in separate papers [9,10].

3.2. Partic density (PD), bulk density (BD), and total porosity

PD of unwashed ALRP (Table 2) represents an average of the densities of its soluble (2.165 Mg m^{-3} for NaCl) and insoluble components. Washed ALRP typically had a PD of $3.05\text{--}3.08 \text{ Mg m}^{-3}$ ($\text{Al} = 2.69$, $\text{Al}_2\text{O}_3 = 3.965$, $\text{MgAl}_2\text{O}_4 = 3.60$, $\text{SiO}_2 = 2.65$, and $\text{CaF}_2 = 3.18 \text{ Mg m}^{-3}$ [17]), whereas unwashed ALRP had a PD ranging from 2.63 to 2.94 Mg m^{-3} . Average PD of FGDP was 2.31 Mg m^{-3} . Particle densities of unwashed

ALRP–FGDP ($2.31\text{--}2.67\text{ Mg m}^{-3}$) were between the values for NaCl (2.165 Mg m^{-3}) and unwashed ALRP (on average 2.77 Mg m^{-3}). The average value was 2.46 Mg m^{-3} . Particle densities of the main components of natural soils typically range from 2.5 to 2.8 Mg m^{-3} [18].

BD ranged from 0.56 Mg m^{-3} for washed FGDP to 0.93 Mg m^{-3} for ALRP (Table 2). High and low values within the group of unwashed materials corresponded to the respective PD values, suggesting that PD and percentage of primary particles, in particular salt content, rather than differences in structure were responsible for the observed BD values. BD of unwashed ALRP ranged between 0.88 and 0.99 Mg m^{-3} . Mineral soils typically have bulk densities of $1.2\text{--}1.8\text{ Mg m}^{-3}$ [34].

Average total porosity of ALRP, FGDP, and ALRP–FGDP was high, ranging from 63.0 to 70.6% for unwashed material and from 67.2 to 75.7% for washed material (Table 2). This resulted in a low BD, even when the PD was high (see Eq. (2)). Provided that the pores are well interconnected, a high total porosity should make ALRP–FGDP a useful material for the uptake and storage of infiltrating rainwater that is to be withheld from seeping into the underlying potash mine residue (NaCl). Also, after salts have been leached from the mix, it should provide physical properties that are favorable for root penetration.

3.3. Particle-size analysis

Particles of unwashed ALRP were separated according to size limits by wet sieving and sedimentation. Both procedures involve intensive mixing of sample solids with water. Salts were leached, and the influence of solid salts on particle size distribution therefore could not be assessed. Material loss during the analysis was recorded and was assumed to equal initial salt content. Salt contents were within 4% of those determined from Eq. (1), see Table 2. Particle-size distribution in Table 3 is listed as percentage by weight of dry, salt-free (or washed) samples. The texture of both washed ALRP and FGDP was silt loam, and accordingly was the texture of ALRP–FGDP.

Table 3
Particle-size distribution of washed ALRP, FGDP and washed ALRP–FGDP (excluding salts)

Class (mm)	ALRP (wt.%) ^a		FGDP (wt.%) ^b	ALRP–FGDP (wt.%) ^c
	Mean	S.D.	Mean	Mean
<0.002	9.0	2.60	25.3	15.0
0.002–0.0063	16.5	3.37	12.7	15.1
0.0063–0.02	19.3	3.28	29.2	22.6
0.02–0.063	17.5	3.48	22.8	19.2
0.063–0.2	22.6	4.12	9.9	18.0
0.2–0.63	13.0	4.52	0.1	8.8
0.63–2.0	2.1	1.66	0.0	1.3

^a By sieving and sedimentation ($N = 10$).

^b By laser-light scattering; data provided by the GfR Company.

^c Computed from mean values for ALRP and FGDP, using mixing fractions (5.2:3) as weighting factors.

Table 4

Apparent intrinsic permeability to salt solution (k_s) and pure water (k_w), as well as saturated hydraulic conductivity (Ks) to salt solution (K_{s_s}) and water (K_{s_w}) of unwashed and washed ALRP, FGDP, and ALRP–FGDP ($N = 3$)

Material	k_s		K_{s_s}	
	Mean (μ^2)	Range (μ^2)	Mean (cm d^{-1})	Range (cm d^{-1})
<i>Unwashed</i>				
ALRP	4.5	4.48–4.53	293.8	284.7–305.2
FGDP	0.9	0.80–1.02	74.7	70.1–78.9
ALRP-FGDP	2.2	1.90–2.45	155.5	145.4–171.2
<i>Washed</i>				
	k_w		K_{s_w}	
ALRP	0.1	0.09–0.14	11.2	9.5–12.4
FGDP	0.3	0.23–0.31	21.6	18.4–25.4
ALRP-FGDP	0.1	0.05–0.11	6.7	5.8–7.3

3.4. Saturated hydraulic conductivity and Intrinsic permeability

Densities and dynamic viscosities of aqueous solutions in equilibrium with ALRP, ALRP–FGDP, or FGDP (all unwashed) were 1.15, 1.12, and 1.01 Mg m^{-3} , and 1.51, 1.36 and 1.05 mPa s, respectively. To minimize the effect of fluid density and viscosity on permeability, and to make results comparable, intrinsic permeabilities were calculated from Eq. (3). Results are given in Table 4.

Saturated conductivity of unwashed samples showed highest values for ALRP (294 cm per day), lowest for FGDP (75 cm per day), and intermediate values for the mix (156 cm per day, Table 4). This ranking is in agreement with intrinsic permeabilities as determined with the salt solution. Measured values were similar to values often found for fine sand [23], although particle distribution was a silt loam. Permeabilities and saturated hydraulic conductivities were higher for unwashed samples than for washed samples. This does not correspond to porosities, which were found to be higher for washed samples. An explanation for this behavior might be that space, initially taken up by solid, but soluble salts in dry unwashed samples, became available to solute movement after saturation. It seems that solid salts lead to an underestimation of original porosity.

Also, permeability and hydraulic conductivity are related to pore-size distribution, pore geometry, and continuity. Thus, they are often inversely related to total porosity. For example, holding structure constant, clayey structured soils have much higher total porosity values than sandy textures, but much lower permeability and conductivity values due to restrictive pore-size. High permeability and Ks values for unwashed materials may possibly be due to the effect of high salt content on soil structure. It is likely that the high ionic concentrations in the unwashed ALRP promoted the flocculation of fine particles resulting in an increase in pore-size and continuity.

3.5. Moisture retention

Average water content at saturation was not determined from measurements, but calculated from porosity. Following [24], it was assumed that this water content was 7.5%

Table 5

Moisture retention, water-holding capacity, and van Genuchten parameters of unwashed and washed ALRP, washed FGDP, and washed ALRP–FGDP ($N = 3$, unless noted otherwise)

	Unwashed		Washed					
	ALRP ^a		ALRP		FGDP		ALRP–FGDP	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Suction (m)	Volumetric water content (%)							
0.0 ^b	58.9	–	62.3	–	68.2	–	59.7	–
0.1	61.3	55.3–69.2	64.6	64.0–65.0	72.1	71.7–72.5	65.0	64.8–65.1
0.32	ND	ND	62.6	61.3–63.6	69.3	69.1–69.8	63.6	63.3–63.9
0.6	48.5	40.9–62.9	61.3	60.8–62.0	68.2	67.8–68.7	62.4	62.1–62.9
1.0 ^c	44.5	35.4–57.4	60.6	60.0–61.2	67.7	67.3–67.5	61.8	61.2–62.5
3.0	31.0	21.8–44.4	59.1	58.2–60.4	62.4	61.7–63.8	60.8	60.0–61.8
158.49	ND	ND	24.3	24.0–24.7	12.0	11.5–12.2	16.8	16.3–17.1
	van Genuchten parameters							
θ_r (%)	17.0	–	0.0	–	0.0	–	0.0	–
θ_s (%)	60.3	–	62.8	–	69.7	–	62.6	–
α (m ⁻¹)	1.682	–	0.148	–	0.180	–	0.070	–
n	1.674	–	1.299	–	1.523	–	1.543	–
m	0.403	–	0.230	–	0.346	–	0.352	–

^a $N = 6$.

^b Volumetric water content at 0.0 m of suction was set equal to porosity calculated with minus 7.5%, using average values for PD from Table 2, and oven-dried weights from retention measurements.

^c Water-holding capacity.

smaller than porosity because of entrapped or dissolved air. Values ranged from 58.9 to 68.2% (Table 5), the lowest value being that of unwashed (saline) ALRP.

For salt-free samples, the 1.0 m suction water content (representing field capacity, which is found in natural soils after gravity drainage has occurred following rain) was on average only 3.9% less than the 0.1 m water content (Table 5). For unwashed ALRP, the difference was 16.8%. There was considerably less water in washed materials at the permanent wilting point (158 m of suction) than at 3.0 m of suction (Table 5), indicating that a high percentage of water was held in small pores, or thin water films with little or no liquid continuity. The 158 m suction water content was highest for washed ALRP (24.3%).

Eq. (4) was used to calculate retention curves from the van Genuchten parameters given in Table 5. The quantities θ_r and θ_s are the fitted residual and saturated water contents, respectively. As starting values for θ_s and θ_r in the fitting procedure with the RETC code, water contents at 0.0 and 158.49 m of suction from Table 5 were selected. Values of r^2 for regressions of observed versus fitted values varied from 0.987 to 0.995. Retention curves from fitted parameters given in Table 5 are shown in Fig. 1.

Water retention curves in Fig. 1 reveal similar hydraulic behavior for all washed (salt-free) materials. The finest-grained material (FGDP) had the largest water content at low suctions, but the lowest water content at high suctions. Washed ALRP had a lower water content at low suctions, a higher water content at high suctions, and a similar bubbling pressure as FGDP. ALRP–FGDP was intermediate. The unwashed-ALRP retention curve is

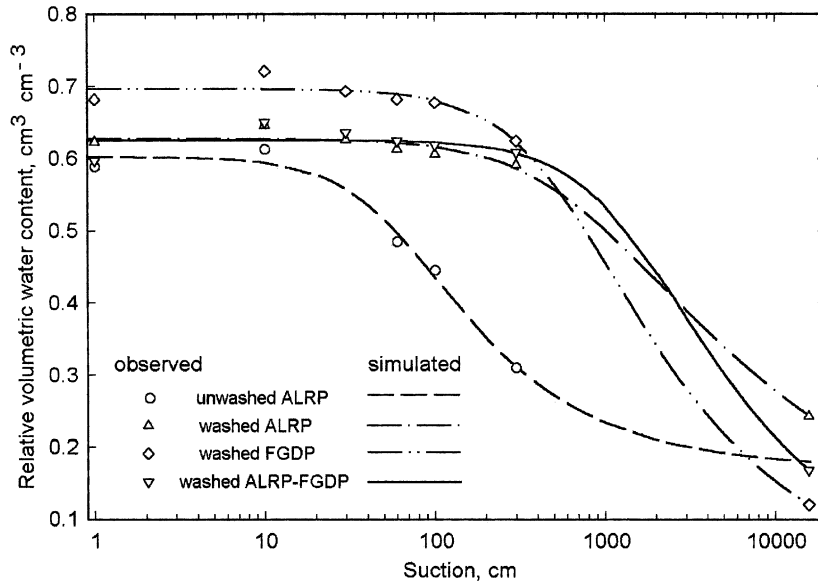


Fig. 1. Moisture retention curves for unwashed and washed ALRP, and for washed FGDP and ALRP-FGDP. Measured data and van Genuchten parameters for the fitted curves are given in Table 5.

significantly different from the washed-material curves in that it has a lower water content at low suctions and a lower air entry value.

Washed samples of all materials did not reveal visible signs of swelling or shrinking as is often observed with fine textured soils when clay particles expand or contract as the result of an uptake or loss of water from interlayer clay particles. This indicates that the material structures were relatively rigid, meaning that a decrease of water content required the entry of air. With unwashed ALRP samples, settling occurred during saturation and dewatering, and totaled 23.3% of sample volume. Because nonsoluble ALRP compounds (i.e. washed ALRP) are rigid, there was no reason to assume that the volume loss of unwashed ALRP was the result of shrinking due to dewatering. Therefore, loss of sample volume most likely was due to dissolved and leached salts. Volumetric water content at different suctions was corrected for loss in total volume, assuming that salt loss was not larger than the visible loss of volume, i.e. assuming that the porosity remained constant. However, no measurements were conducted to test this assumption and the water retention curve for unwashed ALRP, shown in Fig. 1 (and volumetric water contents given in Table 5), might not be totally free from artefacts.

3.6. Consistency, erodibility, and erosion

The Atterberg limits (liquid limit and plastic limit) and the plasticity index are measures of the ability of a porous material to be deformed across a range of moisture contents. The liquid limit is the gravimetric moisture content at which a material will flow when subjected

Table 6
Atterberg limits and plasticity index of unwashed and washed ALRP, FGDP, and ALRP–FGDP

Material	Liquid limit (%) moisture ^a	Plastic limit (%) moisture ^a	Plasticity index (%)
Unwashed			
ALRP	21.5	19.3	2.2
FGDP	34.8	32.2	2.6
ALRP–FGDP	22.3	19.5	2.8
Washed			
ALRP	54.1	47.6	6.5
FGDP	52.8	43.2	9.6
ALRP–FGDP	43.2	34.2	8.9

^a Gravimetric moisture content.

to a striking force. It is an indirect measurement of shear strength. The higher the moisture content at the liquid limit, the lower the potential of the material to flow or slump under normal moisture conditions. Data are given in Table 6.

Unwashed ALRP–FGDP samples had an average liquid limit of 22.3% moisture by weight. Because the moisture content of the material that is leaving the mixer is about 13%, it does not slump under normal moisture conditions. However, the liquid limit helps to explain why initially ALRP, dewatered under pressure to a moisture content of 35%, could not be transported upslope on a conveyor belt. The liquid limit of FGDP (34.8%) was well above the average moisture content of this material when it entered the mixer (<3.0%). Water content of all materials at the liquid limit was lower than water content at 1 m of suction (\approx field capacity, see Table 5). After heavy rainfall, this may potentially lead to stability problems, because material may slump before it will be dewatered by gravity flow.

The plastic limit is the gravimetric moisture content of a material at which it will begin to exhibit plastic behavior. The higher the moisture content at the plastic limit, the less plastic the material will be at normal moisture contents. The plasticity index, the range of moisture contents over which the material is plastic, was only 2.2% for unwashed ALRP and 2.8% for unwashed ALRP–FGDP. This indicates that both materials change from nonplastic, via plastic, to liquid behavior over a narrow range of water contents. Adding dry FGDP to ALRP, dewatered in the decanting centrifuge to 18%, resulted in an average moisture content of 13% of the mix (see earlier section). This seems to indicate that the positive effect of adding FGDP was due partly to a reduction of the initial moisture content of ALRP. The result was a nonplastic, granular material. On the test site at the Sigmundshall waste mound it was observed that this material (ALRP–FGDP) flowed smoothly down the long slope, thus yielding an evenly thick surface cover.

Liquid limit, plastic limit, and plasticity index of all materials were considerably higher after washing (Table 6). This suggests that the stability of the proposed cover layer material will increase with decreasing salt content over time under the ambient humid climate. Chemical reactions in the moistened FGDP, similar to those occurring in gypsum or hydraulic cement, may also contribute to an increase in stability. However, such reactions were not part of the research conducted during this study.

With the rainfall erosivity index relation given by [30], the factor R in Eq. (5) was estimated as $R = 51.4 \text{ kJ mm m}^{-2} \text{ h}^{-1}$, for a mean annual rainfall at the site of 640 mm. For the erodibility factor of pure ALRP we calculated $K = 0.65 \text{ Mg ha}^{-1} \text{ N}^{-1}$. Because the length of the relevant slope of the mound was 130 m and the mean angle of this slope was 36° , the topographic factor in Eq. (5) was calculated as $LS = 28.7$. For $C = 1$ and $P = 1$, a value for the average annual ALRP loss due to erosion by water was calculated as $A = 959 \text{ Mg ha}^{-1}$. When compared with agricultural soils exposed to similar conditions, both the calculated K and A value are high. It is obvious that these calculations can only give a rough estimate about the erodibility and the possible erosion losses of ALRP as a cover material. However, the computational results were confirmed by observations during the initial stage of the project, when pure ALRP was used as cover material at the test site and considerable erosion was noticed. Therefore, ALRP was mixed with FGDP, and with an ALRP–FGDP cover layer, erosion practically ceased to exist. A possible reason is the formation of carbonate and silica cement in the mix. Fly ash materials, such as FGDP, frequently undergo pozzolonic reactions, which have a strong erosion-reducing effect (the name is derived from Pozzuoli, a city near Naples, Italy, where since Roman times it has been known that volcanic ashes, mixed with burned limestone, yield strong mortar and cement). For ALRP–FGDP no K or A value was calculated, because RUSLE does not consider pozzolonic effects.

4. Conclusions

The objective of our study was to determine some selected physical properties of a fine-grained, saline industrial by-product (ALRP). This by-product from the aluminum recycling industry cannot be disposed of in landfills, because of its high salt (NaCl) content. Therefore, the waste has been proposed to serve as a soil substitute in a surface barrier over a potash mining residue mound. This mound itself consists almost entirely of NaCl and produces therefore large amounts of briny runoff during the wet winter season. Covering the mound with a layer of soil-like ALRP would relieve the environment in two ways; it would reduce both the mound's runoff and the waste's landfill disposal problem. It is likely that after most of the salt (NaCl) has been leached from the barrier, this surface layer is able to support vegetation, to enhance evaporation, and to reduce the amount of briny runoff from the potash mining residue mound. To judge the geotechnical feasibility of the waste for this purpose, its properties, in particular its physical properties such as texture, porosity, and water-holding capacity, must be known.

Physical properties of ALRP, mainly from a metallurgical point of view, have been reported previously [35]. Our study, however, appears to be the first survey on some of the basic soil physical and hydraulic properties. Generally speaking, our data show that ALRP and ALRP–FGDP have physical and hydraulic properties that resemble those of medium to fine textured soils. This applies in particular to the water retention curves. These curves are similar to those of medium to fine textured soils with predominantly small pores and a medium to high water-holding capacity. Also, it is possible to describe these curves with the van Genuchten model [25]. The unwashed materials showed further a surprisingly high permeability. PD and BD of the various materials appeared to reflect the ratio of the

different components in the mixtures. Of much importance is the erodibility of the studied materials. Apparently because of the cementing effect of FGDP, the ALRP–FGDP cover on the steep slope of the experiment site of the mound did not show noteworthy slumping or runoff-induced erosion. Our data concerning the physical properties, in particular those of the ALRP–FGDP mix, therefore seem to indicate that the mix is suited to serve as cover material, even on steep slopes of high salt mine residue mounds. The use of ALRP–FGDP as a cover material for potash mining residue mounds could also help solve the ALRP landfill disposal problem. The collected data in this study may serve future users of similar products.

From a physical point of view it thus seems possible to use ALRP as a soil-like substitute in a surface barrier, especially when it is mixed with the FGDP. Because the initial washout of salt does not worsen the environmental impact of the bare potash mining residue mound (which itself produces large amounts of briny runoff) its use as a soil-like substitute is environmentally meaningful. After the initial salt content is washed out and vegetation has been established, the surface barrier helps to reduce considerably the amount of briny discharge from the residue mound. The use of ALRP as a surface barrier material thus makes double sense: on one hand it helps to reduce the annual amount of briny discharge from the potash mining residue mound and on the other hand it helps to reduce the problem of disposing hazardous industrial wastes.

Additional work on the feasibility of ALRP as a surface barrier material includes a lysimeter study to observe and to model water and salt dynamics of the material under field conditions. For details the reader is referred to Hermsmeyer et al. [9,10].

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