

An Eco-Innovative Solution for Reuse of Leachate Chemical Precipitation Sludge: Application to Sanitary Landfill Coverage

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ABSTRACT

Sanitary landfill leachate was treated by chemical precipitation and the potential use of mixtures of the chemical precipitation sludge (CPS) generated and natural soil to cover the solid waste at sanitary landfills was evaluated. Tests were performed with soils having 0, 5 and 10% CPS, and the organic matter, pH, compaction, hydraulic conductivity, particle-size, liquid limit, plastic limit, and plasticity index were determined. It was found that leaching increased with CPS concentration, as well as the fine particles content in the soil, which reduced the hydraulic conductivity. pH was the most affected parameter, with values of 7.03, 7.12, and 11.46 for 0, 5 and 10% CPS, respectively. However, at 5% CPS, biodegradability was favored without a significant increase in the leaching process, showing that CPS can be used as temporary cover material, without adversely affecting the landfill system, being an eco-innovative solution for the final disposal of the sludge.

Keywords: sludge reuse, landfill, cover material, chemical precipitation.

INTRODUCTION

Sanitary landfilling is, nowadays, the most applied method for waste disposal. By utilizing the principles of engineering, wastes are reduced to the smallest practical volume and confined to the smallest practical area, which is covered with a layer of soil to avoid damages or hazards to public health or safety [Raghab et al. 2013]. This process generates leachate, representing a major problem for solid waste landfills management. Landfill leachate is the liquid resulting from the natural humidity and water present in wastes, from the organic matter degradation, and from water infiltration in the covering and inner layers of landfill cells [Peng 2013]. It is a dark-colored

liquid with an unpleasant odor and high values of chemical oxygen demand (COD), pH, NH_4^+ , and heavy metals, being the physico-chemical characteristics dependent on the age of the landfill [Raghab et al. 2013].

Conventional landfill leachate treatment processes include physicochemical, biological and membrane technologies, generating sludge as the usual by-product of these treatments. This sludge contains a large quantity of pollutants, which may cause secondary pollution to the environment if handled improperly [Wang et al. 2010, Jun et al. 2015]. Therefore, sludge management is of extreme importance and should be a fundamental part of any landfill leachate treatment plant [Xu et al. 2014].

For sanitary landfilling process, a large amount of cover materials is required to cover the wastes. Soil mixed with sludge can be an alternative to traditional cover materials. In fact, the pollution caused by the sludge can be prevented in a closed landfill system [Jun et al. 2015]. Typically, cover systems can be divided into temporary cover (including daily cover and interim cover) and final cover. The main purpose of the temporary cover is to reduce the rain infiltration and prevent the wastes from being exposed to rain [Kamon et al. 2002]. Figure 1 shows the construction criteria for the cover systems recommended by the United States Environmental Protection Agency, which states that the recommended hydraulic conductivity in barrier layer in a cover system for a municipal landfill must be less than or equal to $1 \times 10^{-5} \text{ cm} \cdot \text{s}^{-1}$ [Kamon et al. 2002].

Li et al. (2003) and Jun et al. (2015) suggested that the temporary cover in a sanitary landfill should enable the leachate recirculation, ensuring, at the same time, uniform degradation and waste stability. Alternative daily cover materials are suggested in the Standard Guide for Evaluation and Selection of Alternative Daily Covers for Sanitary Landfills [ASTM 2005], which includes foams, spray-on slurries, geosynthetics, and indigenous materials (such as sludge, ash, and shredded tires), with different levels of permeability [Jun et al. 2015].

The use of dewatered sludge as temporary landfill cover material is also an option that has been widely studied [Jiao 2007, Zhou and Wu 2011, Chen et al. 2013]. Still, there is a lack of deep understanding of the interaction between the soil, the sludge, and the rain water [Li 2006, Yang et al. 2012]. During the operation and after the closure of the sanitary landfill, the temporary cover is soaked into the leachate.

The aim of the present study was to evaluate the possibility of using chemical precipitation sludge (CPS) mixed with natural soil as an aggregate for temporary cover material at a sanitary landfill. The CPS was obtained from a basic chemical precipitation (BCP) treatment of the sanitary landfill leachate. The effectiveness of the CPS reuse was accessed and discussed.

MATERIALS AND METHODS

The sanitary landfill leachate samples were collected from the entrance of a stabilization lagoon at the non-hazardous waste sanitary landfill of Association of Municipalities of Central Alentejo (AMCAL), located in Vila Ruiva, Cuba Municipality, Beja District, Portugal.

The leachate sample was treated by a basic chemical precipitation process [Ramalho 2015], through addition of 160 mL of a $200 \text{ g} \cdot \text{L}^{-1}$ aqueous calcium oxide solution to 1 L of leachate, at constant stirring of 300 rpm, for 40 min. A sedimentation period of two hours followed precipitation, to separate sludge from the supernatant. The obtained CPS was then dried under ambient conditions to dehydrate. CPS was characterized through electrical conductivity, pH, total solids, volatile solids, total Kjeldahl nitrogen (TKN), and NH_4^+ -N determinations, according to the Standard Methods for the Examination of Water and Wastewater [APHA 2012].

CPS was mixed with the soil utilized as temporary cover material at the sanitary landfill of AMCAL. Different CPS mass concentrations were studied, namely 0, 5 and 10%.

Surface soil sampling was performed in accordance with the ISO 18400-101:2017 standard [ISO 2017]. The soil was classified according to the Unified Soil Classification System using the

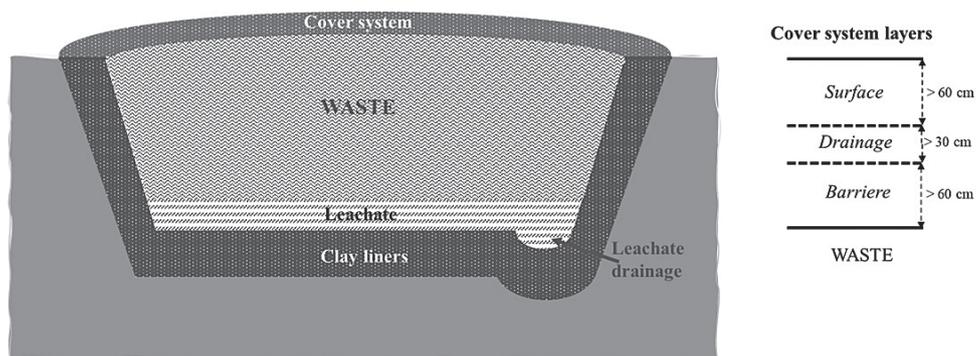


Fig. 1. Construction criterion for cover systems recommended by USEPA (adapted from Kamon et al. 2002)

Standard Practice for Classification of Soils for Engineering Purposes ASTM D2487 [ASTM 2017a], based on laboratory determinations of particle-size characteristics, liquid limit, plastic limit, and plasticity index. Grain size distribution was determined according to the Standard Test Method for Particle-Size Analysis of Soils ASTM D-422 [ASTM 2007]. The determinations of liquid limit, plastic limit, and plasticity index, were performed according to the ASTM D4318-10 standard [ASTM 2010]. The organic matter content was determined by the Walkley and Black method [Galantini et al. 1994]. The pH was determined according to the EPA 9045D method, outlined by the United States Environmental Protection Agency [U.S. EPA 2004].

The compaction and hydraulic conductivity characteristics of the soil and soil+CPS mixtures were also determined. The compaction method, used to determine the relationship between molding water content and dry unit weight of soils, followed the ASTM D698-12e2 standard, using the Standard Effort [ASTM 2017b]. The hydraulic conductivity of the saturated porous was analyzed according to the ASTM D 5084-03 standard [ASTM 2016]. Three replicates were performed for each determination.

In order to evaluate the possible mobility of contaminants, present in the cover material by the introduction of CPS, the leaching tests were performed based on the Standard EPA Test Method 1312 [U.S. EPA 1994]. Batch-type experiments were run at laboratory scale, using ASTM type II reagent water as extraction fluid, for a period of 48 hours of leaching. The liquid phase was analyzed through pH, electrical conductivity, oxidation-reduction potential, absorbance at 254 and 410 nm, COD, biochemical oxygen demand (BOD_5), total and calcium hardness, total and phenolphthalein alkalinity, Cl^- , SO_4^{2-} , and NH_4^+ -N determinations, performed in triplicate. All the determinations were performed according to the

Standard Methods for the Examination of Water and Wastewater [APHA 2012].

RESULTS AND DISCUSSION

Table 1 presents the characterization of the CPS obtained from the sanitary landfill leachate basic chemical precipitation treatment. The high pH value presented was due to the calcium hydroxide formed by the reaction of calcium oxide with water in the BCP process. The electrical conductivity result indicated the CPS high salinity. The relationship between NH_4^+ -N and TKN was 7.5% and, when compared to the same relationship for the raw leachate (88%), suggests a good NH_4^+ -N removal during BCP.

The soil sample from the AMCAL landfill site, utilized in this study for CPS+soil mixtures, was composed of 65.71% gravel, 22.95% sand, 6.94% silty and 4.4% clay. According to the Unified Soil Classification System [ASTM 2017a], the soil used in the present study is classified as clayey gravel with sand, gravel-sand-clay mixtures, group symbol GC. Particle-size distribution of the analyzed sample is presented in Figure 2 and the soil analysis is presented in Table 2. An important fact to highlight is the absence of organic matter in the soil. Additionally, the pH value indicates a neutral soil.

For the different CPS concentrations utilized, the optimum moisture content to achieve the maximum density, resulting from the compaction tests, were $8.6 \pm 0.1\%$, $9.9 \pm 0.2\%$ and $10.8 \pm 0.2\%$ for 0, 5 and 10% CPS, respectively (Fig. 3). Regarding the hydraulic conductivity of the compacted samples with the optimum moisture content (Proctor test, for maximum density), the obtained results were 4.6 ± 0.1 , 3.2 ± 0.1 and $1.3 \pm 0.1 \times 10^{-6} \text{ cm} \cdot \text{s}^{-1}$, for 0, 5 and 10% CPS, respectively. The increase in CPS concentration increases the content of fine particles in the soil, increasing the optimum

Table 1. Characterization of the chemical precipitation sludge used in the study

Parameter	Mean value \pm Standard deviation
pH	12.40 \pm 0.02
Electrical conductivity, $\text{mS} \cdot \text{cm}^{-1}$	28.5 \pm 0.2
Total solids, %	98 \pm 1
Volatile solids, %	11 \pm 1
Total Kjeldahl nitrogen, $\text{g} \cdot \text{kg}^{-1}$	2.0 \pm 0.2
$N-NH_4^+$, $\text{g} \cdot \text{kg}^{-1}$	0.15 \pm 0.02

Table 2. Characterization of the soil used as daily cover at AMCAL sanitary landfill site

Parameter	Mean value
pH	6.9
Organic matter	0.1%
Liquid limit	32%
Plastic limit	23%
Plasticity index	9%

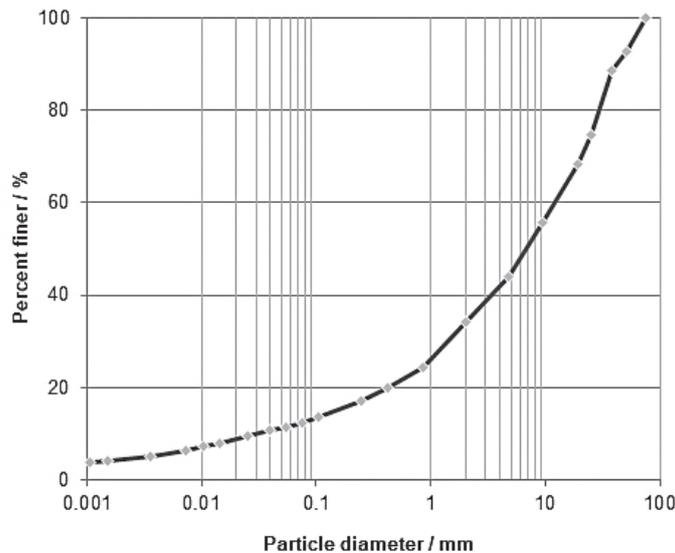


Fig. 2. Particle-size analysis of the soil used as daily cover at AMCAL sanitary landfill site

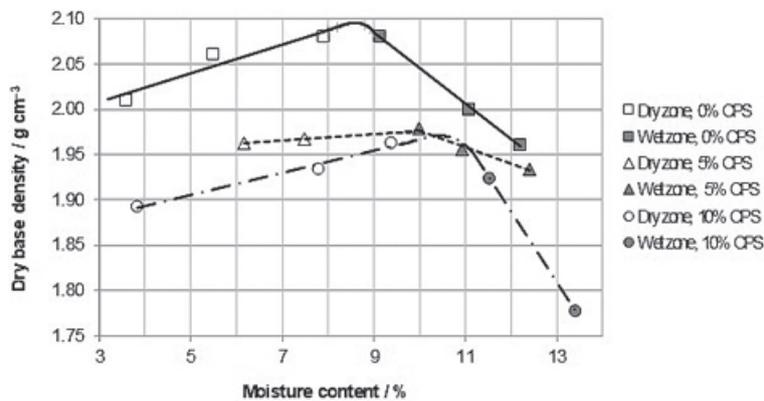


Fig. 3. Compaction test results obtained for the different CPS concentration in soil

moisture content and reducing the hydraulic conductivity, as reported by Rico and del Castillo (1992). For all the CPS concentrations studied, the hydraulic conductivity results were within the limits recommended by USEPA, with values below $1 \times 10^{-5} \text{ cm} \cdot \text{s}^{-1}$ [Kamon et al. 2002].

The addition of CPS to the soil increases the content in particles with dimensions smaller than 0.002 mm, allowing a similar effect to the clay, which is assumed to have a significant capacity to improve plasticity and soil compaction conditions which act in the same way as clay, with a significant capability for modifying the compaction conditions of the soil [Safari and Bidhendi 2007]. Compaction is a mechanical process that, through repeated and rapid application of loads to the soil, leads to a decrease in its volume and, consequently, to a decrease in the void index and an increase in dry weight. This reduction in volume results essentially from the expulsion of air from the voids of the soil, without any significant

change in the water content or change in the volume of the solid particles during the process. When applying a particular compaction energy, the resulting specific mass is a function of the moisture content that the soil contains. When compacted with a low moisture content, the friction between particles is very high and no significant void reduction is achieved. In compaction with higher moisture contents, water acts as a lubricant between the particles that slide between them, accommodating themselves in a more compact arrangement.

The mixtures of soil+CPS, with different CPS contents were submitted to leaching tests and results are presented in Table 3. The addition of CPS to the soil resulted in a pH increase in the resulting leachate, mainly at 10% CPS. The negative value found for the oxidation-reduction potential, at 10% CPS, is consistent with a reducing medium, which is alkalized by the generation of OH^- ions. Reactions such as the reduction of

Table 3. Results of the leaching tests utilizing different CPS concentrations

Parameter	CPS concentration in soil		
	0%	5%	10%
pH	7.03 ± 0.03	7.12 ± 0.03	11.46 ± 0.07
Oxidation-reduction potential, mV	186 ± 4	172 ± 2	-25 ± 1
Absorbance at 254 nm (dilution 1:50)	0.012 ± 0.002	0.017 ± 0.001	0.032 ± 0.001
Absorbance at 410 nm	0.066 ± 0,001	0.071 ± 0,001	0.093 ± 0.001
Chemical oxygen demand, mg·L ⁻¹	245 ± 2	263 ± 8	498 ± 2
Dissolved chemical oxygen demand, mg·L ⁻¹	68 ± 1	104 ± 7	374 ± 4
Biodegradability index	0.11	0.21	0.13
Biochemical oxygen demand, mg·L ⁻¹	27 ± 2	56 ± 2	67 ± 3
Total alkalinity, mg·L ⁻¹ CaCO ₃	238 ± 8	340 ± 9	626 ± 13
Phenolphthalein alkalinity, mg·L ⁻¹ CaCO ₃	Not detected	Not detected	408 ± 11
Cl ⁻ , mg·L ⁻¹	15 ± 3	122 ± 2	460 ± 5
SO ₄ ²⁻ , mg·L ⁻¹	11.7 ± 0.2	15.1 ± 0,1	27.1 ± 0.2
NH ₄ ⁺ -N, mg·L ⁻¹	0.19 ± 0.08	0.23 ± 0.08	0.9 ± 0.2
Electrical conductivity, mS·cm ⁻¹	232	513	2165
Total hardness, mg·L ⁻¹ CaCO ₃	47.1	94.2	306.1
Calcium hardness, mg·L ⁻¹ CaCO ₃	19	57	254

sulfate and the generation of methane, both wanted in the landfilling process, are favored under these conditions. The absorbance results at 410 nm indicate an increase in color intensity with CPS concentration. Similarly, the content in high molecular weight compounds, with high degree of aromaticity and phenolic groups, directly related with the absorbance at 254 nm, increased with CPS concentration. In fact, all the analyzed parameters increased their values with CPS concentration, mainly when CPS concentration was changed from 5 to 10%.

CPS addition to the soil increases the electrical conductivity and the total and calcium hardness. Again, this increase is more pronounced when the CPS concentration is raised from 5 to 10%. The increase in hardness can be attributed to the calcium present in the CPS, from the calcium hydroxide addition during the chemical precipitation process. The increase in electrical conductivity can also be explained by the presence of calcium, as well as other ionic species present in CPS, such as chloride and sulfate.

Regarding the biodegradability index (ratio between BOD₅ and COD, BI) of the leachates obtained from the leaching tests, the highest BI was found for the mixture soil + CPS-5%, with an increase of 91% compared to the soil without sludge. This result indicates that the addition of CPS at 5% has a positive effect on the biodegradability of the leachates that will

be generated in the landfill, favoring the landfill system.

Attending to the results described, it can be concluded that CPS can be successfully employed as an aggregate for the cover material in landfill sites. Although there is an increase in pH and dissolved solids, there is no negative impact on the landfill operation.

The AMCAL landfill produces, during rainfall season, up to 50 m³ per day of leachate [Figueira 2009]. Considering that each liter of treated leachate generates 27.8 g of CPS [Ramalho 2015], approximately 1390 kg of CPS would be produced per day. Applying a mixture of soil + CPS-5% would lead to, approximately, 27,800 kg of cover material per day, which, according to Ramalho (2015), ensures the total integration of the sludge produced in to the landfill system, providing an innovative alternative for its use.

CONCLUSIONS

The soil used in this study as covering material was clayey gravel with sand, with a low hydraulic conductivity when compacted ($\leq 1 \times 10^{-5}$ cm·s⁻¹), according to the recommended for a correct sanitary landfill operation. The addition of CPS to the soil used as cover material increased the pH and the optimum compaction humidity. However, this condition is compensated by the

reduction in the hydraulic conductivity, so that the increase in humidity will not affect the correct operation of the landfill due to runoff, although it is necessary to have a good system drainage on the surface to avoid waterlogging.

CPS addition to the cover material also led to an increase of fine material, which acts in the same way as if it was clay. It assumes a structure in the form of panels, with hollow stacks between them, allowing the compaction.

The mixture of soil+CPS-5% presented the best results. It led to an increase of the biodegradability of the leachate, in the leaching test, without increasing the leaching process, as occurred in the case of the mixture of soil+CPS-10%.

The use of sludge as an aggregate for the temporary cover material does not negatively affect the landfill and can prevent the pollution problems caused by the sludge management. Thus, the addition of CPS to the soil usually utilized as cover material is an eco-innovative and environmentally friendly option that avoids poor sludge management and adverse effects of the landfill system.

Acknowledgments

The authors gratefully acknowledge the financial support received from Fundação para a Ciência e a Tecnologia, FCT, through the funding of the UID Fiber Materials and Environmental Technologies (FibEnTech), project UIDB/00195/2020, and the contract funding awarded to A. Fernandes.

REFERENCES

1. APHA 2012. Standard Methods for the Examination of Water and Wastewater, 12th ed. American Public Health Association, American Water Works Association and Water Environment Federation, Washington.
2. ASTM 2017a. Standard Practice for Classification of Soils for Engineering Purposes (Unified Soil Classification System) Standard ASTM D2487, in: ASTM Volume 04.08 Soil and Rock (I): D421-D5876.
3. ASTM 2017b. Standard Test Methods for Laboratory Compaction Characteristics of Soil Using Standard Effort (12 400 ft-lbf/ft³ (600 kN-m/m³)), in: ASTM Volume 04.08 Soil and Rock (I): D421-D5876. West Conshohocken.
4. ASTM 2016. Standard Test Methods for Measurement of Hydraulic Conductivity of Saturated Porous Materials Using a Flexible Wall Permeameter 1.
5. ASTM 2010. Standard Test Methods for Liquid Limit, Plastic Limit, and Plasticity Index of Soils D4318-10. Report 4, 1-14.
6. ASTM 2007. Standard test method for particle-size analysis of soils: ASTM D 422. ASTM International, 63, 1-8.
7. ASTM 2005. Evaluation and selection of alternative daily covers (ADCs) for sanitary landfills (D6523).
8. Chen P., Lin W.A., Zhan X.J., Yiang Y.Y. 2013. Influence of leachate soaking on strength and permeability of deeply dewatered. *Rock and Soil Mechanics*, 34, 337-341.
9. Figueira C.M. 2009. Evaluation of the operation of the leachate treatment station at the Villa Ruiva landfill and rehabilitation proposal. M.S. Dissertation, Faculty of Science and Technology, New University of Lisbon, Lisbon, Portugal.
10. Galantini J., Rosell R., Iglesias J. 1994. Determination of organic matter using the method of Walkley and Black in granulometric fractions of the soil. *Soil Science*, 81-83.
11. ISO 2017. Soil Quality - Sampling. ISO 18400 101:2017. International Organization for Standardization.
12. Jiao Y.J. 2007. Study of sewage sludge of chongqing used as daily cover material in MSW landfill. MSc. Dissertation, Chongqing University, Chongqing, China.
13. Jun H., Feng L., Yong L., Xi-Lin C. 2015. Modified sewage sludge as temporary landfill cover material. *Water Science and Engineering*, 8, 257-262.
14. Kamon M., Inazumi S., Rajasekaran G., Katsumi T. 2002. Evaluation of waste sludge compatibility for landfill cover application. *Soils and Foundations*, 42, 13-27.
15. Li B., Liu D., Yang L. 2003. Selection of temporary cover materials for bioreactor landfills. *Environmental Science & Technology*, 26, 29-30.
16. Li L. 2006. Study on sludge solidification technology and heavy metal pollution control. Ph.D. Thesis, Hohai University, Nanjing, China.
17. Peng Y. 2013. Perspectives on technology for landfill leachate treatment. *Arabian Journal of Chemistry*, 10(2), S2567-S2574.
18. Raghav S.M., Abd El Meguid A.M., Hegazi H.A. 2013. Treatment of leachate from municipal solid waste landfill. *HBRC Journal*, 9(2), 187-192.
19. Ramalho M.S. 2015. Treatment of leachates by chemical precipitation, carbonation, and phytoremediation tuning. MSc. Dissertation, Beja Polytechnic Institute, Beja, Portugal.
20. Rico A., del Castillo H. 1992. Considerations on compaction of soils in transport infrastructure works. *Ministry of Communications and Transportation of México*, 2, 187-200.

21. Safari E., Bidhendi G.N. 2007. Removal of manganese and zinc from Kahrizak landfill leachate using daily cover soil and lime. *Waste Management*, 27, 1551-1556.
22. U.S. EPA 2004. Method 9045D. Soil and Waste pH. United States Environmental Protection Agency.
23. U.S. EPA 1994. Test Method 1312. Synthetic Precipitation Leaching Procedure, SPLP. United States Environmental Protection Agency.
24. Wang W., Luo Y., Qiao W. 2010. Possible solutions for sludge dewatering in China. *Frontiers of Environmental Science & Engineering*, 4, 102-107.
25. Xu W., Xu J., Liu J., Li H., Cao B., Huang X., Li G. 2014. The utilization of lime-dried sludge as resource for producing cement. *Journal of Cleaner Production*, 83, 286-293.
26. Yang J.K., Yank X., Li Y.L., Zhang M., Li Y., He S. 2012. Dewatering of sewage sludge with skeleton builder and engineering properties of dewatered sludge. *Journal of Wuhan University of Science and Technology*, 35, 133-136.
27. Zhou L.Q., Wu C.L. 2011. Study of modified sludge as landfill cover soil. *Chinese Journal of Environmental Engineering*, 5, 2864-2868.