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Otimização Multi-resposta do processo de estabilização/solidificação de resíduo industrial perigoso

Multi-response optimization of the stabilization/solidification process of industrial hazardous waste

Optimización multi-respuesta del proceso de estabilización/solidificación de residuos industriales peligrosos

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Resumo

A Solidificação/Estabilização (S/S) é uma técnica importante no tratamento de resíduos perigosos, devido ao seu baixo custo e fácil processamento. O objetivo deste trabalho foi otimizar e padronizar o processo S/S em uma empresa brasileira. Para tal, estudou-se a compactação do resíduo perigoso usando misturas de diferentes materiais como agentes solidificantes: solo argiloso, um polímero superabsorvente (polpa de celulose) e pó de lixa. Foi avaliado o efeito da massa dos solidificantes sobre o volume da mistura compactada, umidade final e o custo do processo de S/S empregando um Planejamento Composto Central. Os resultados mostraram um custo mínimo de R$ 192,89/ton de resíduo líquido, usando cerca de 71,69 (ton de solo argiloso)/(ton resíduo), com uma umidade final menor que 20% (b.u.).

Palavras chave: gestão de resíduos, resíduo perigoso, superfície de resposta.

Abstract

Solidification/Stabilization (S/S) is an important technique used in waste treatment, due its low cost and easily processing. The aim of this study was to optimize and standardize the S/S process in a Brazilian company. The packing of the dangerous residue was done using a mixture of different solidifying agents, such as: clayey soil; superabsorbent polymer (SAP)/cellulose pulp (fluff) and sanding powder. It was evaluated the effect of different amount of solidifying agent over the volume of packed mixture, the final moisture and the cost of the S/S process, using a Central Composite Design. The results showed a minimum cost for the S/S operation of R$ 192.89/ton of liquid waste, using about 71.69 tons of clay soil for each ton of untreated residue, allowing a highly packed residue with a moisture up to 20% (w.b.).

Keywords: waste management, hazardous waste, experimental design.

Resumen

La solidificación/estabilización (S/S) es una técnica importante en el tratamiento de residuos peligrosos, debido a su bajo costo y fácil procesamiento. Este trabajo propuso optimizar y estandarizar el proceso S/S en una empresa brasileña. Para este propósito, se estudió la compactación de residuos peligrosos utilizando mezclas de diferentes materiales como agentes solidificantes: tierra arcillosa, un polímero superabsorbente (pulpa de celulosa) y papel de lija. El efecto de la masa de los solidificadores sobre el volumen de la mezcla compactada, la humedad final y el costo del proceso de S/S se evaluó mediante la Planificación del Compuesto Central. Los resultados mostraron un costo mínimo de R$ 192,89/ton del resíduo, utilizando 71,69 ton del la tierra arcillosa por ton del reiduo, lo que permitió un residuo tratado con un alto grado de compactación y humedad final inferior al 20% (b.u.).

Palabras clave: gestión de residuos, residuos peligrosos, planificación experimental.

1. Introdução

The great rate of urban growth, industrialization and consumption have led to a significant increase in solid waste generation (Silva et al., 2020). The management of solid waste (SW) is one of the priority concerns in relation to the environment protection and preservation. Due to the problem of the inevitable generation of waste, studies and research are necessary to know the waste generation profile of each region (Evaristo et al., 2017; Andrade et al., 2019; Borges et al., 2019; Santiago et al., 2019; Tavares et al., 2019; Vilela et al., 2020), in order to create feasible treatments that make waste less harmful to the environment or that allow the reuse of waste (Melchert et al., 2011).

Residues classified as hazardous are those that can offer risks to the environment and society, being necessary a rigorous management (Cullinare et al., 1986; ABNT, 2004). The residue treatment has the purpose to convert the waste in a no harmful to the environment or acceptable for current land disposal requirements (Wiles, 1987). The adopted treatment procedures can reduce the residue volume, toxicity, and mobility, making its recovery, storage and transport easier and safer (Cullinare et al., 1986; ABNT, 2004). However, due to the large variety of hazard residues generated in diverse sectors, there are no established standard treatments, being necessary to research and develop processes suited to the local and economic reality (Melchert et al., 2011).

Solidification/Stabilization (S/S) is one of the most important techniques available for waste treatment, ant and has been widely used in several countries for more than 30 years. The S/S can be used in the treatment of hazardous waste with different characteristics and origin (Oliveira, 2003).

Stabilization is defined as the set of techniques that reduce the hazardousness of waste by converting the contaminants into stable, less toxic or less soluble compounds, without necessarily altering the physical state of the substance. Solidification refers to the process that retains the residue into a structurally stable solid.

The aim of solidification/stabilization process is to keep the hazardous waste components within the allowable limits established by regulations, to produce of a solid material from liquid waste or sludge; to improve the physical and handling characteristics of the waste; to decrease the surface area to inhibit the transfer of the waste contaminants among each other, and reduce the components mobility during the exposure to leaching fluids (Balcan & Kocasoy, 2004).

These techniques are considered as non-destructive techniques of immobilization because the hazardous constituents of the waste are not removed or destroyed, they are only transformed in their physicochemical form more stable and less susceptible to leaching (Wiles, 1987; Brito & Soares, 2009; Barth, 1990).

Among the factors that may influence the selection of the technology, design, implementation, and performance of the S/S process and product, it is important to highlight the importance of choosing the appropriate solidifying agent. It represents a great part of the S/S costs and the incompatibility between the residue and the solidifying agent may inhibit the reactions required for the process (Melchert et al., 2011). The volume occupied by the solidified waste must also be taken into account because it is a variable that composes the total operational cost and that will directly influence the life of the landfill. An interesting alternative to the treatment of hazardous waste would be the use of residues from other industrial sectors as solidifying/stabilizing agents (SSA), since the increase in industrial waste generation has become an urgent problem.

The use of clays in the S/S of hazardous waste has been studied due to its properties as adsorbents, its low cost and the ease of obtaining. The adsorptive properties are a consequence of the colloidal nature of these materials. Other characteristics such as surface area and cation exchange capacity will also influence directly the clay adsorption capability (Almeida Neto et al., 2002; Tomasella et al., 2015).

During the construction of landfills, it is necessary to excavate the land to accommodate the waste in the trenches. Since excavation is costly, waste management companies often sell the dirt excavated at a low price or leave it open to sky, occupying large areas. The residues generated by the wood processing operations are a great inconvenience to the timber industry, among which sanding powder and sawdust should receive more attention. These materials have low density, which demands a high storage area. In addition, sanding powder can be explosive (Callé et al., 2005; Daian & Ozarska, 2009).

Several studies report the use of wood waste absorbing nonpolar substances. Annunciado (2005) reported the high oil sorption capacity of sawdust. This residue reached a sorption percentage of 98%. Furthermore, the particle size of sawdust influenced this process, small particles presented greater sorption capacity. These results lead us to believe that sanding powder may also have great sorption potential of nonpolar compounds. Larous and Meniai (2012) verified in their studies that charred sawdust presents a good adsorption capacity to remove phenols from residual waters.

Superabsorbent polymer (SAP) and cellulose pulp (fluff) are hydrophilic gels that can absorb a large amount of water and aqueous solutions (Ahmed, 2015). The high water absorption occurs due to the presence of a reticulated three-dimensional polymer chain that attracts small polar molecules and retains them. This reticulum avoids the solubilization of the chains, but allows their expansion (Santos, 2015). Although SAP has a higher absorption capacity than fluff pulp, it is still necessary to use this fiber in hygienic products because its fibers generate a surface tension that increases the absorption and retention of liquids, besides avoiding direct contact between the polymer and skin, which can cause health issues, as allergies (Vidal & Hora, 2014). The absorption capacity of the fluff fibers can be attributed to the removal of the more hydrophobic component, lignin, during its process of fabrication (Hubbe et al, 2013). Santos (2015) described the use of SAP in water treatment, and showed that SAPs can be applied for the removal of water in aqueous waste. Davies et al. (2004) proposed the detoxification of olive mill wastewater using two superabsorbent polymers. Olive mill wastewater is a problem due to its high content of polyphenols and organic matter. The studies showed that the use of the superabsorbent polymers allowed the use of the water as fertilizer since they immobilized the contaminants reducing their phytotoxicity, and provide to the plant the organic and mineral content necessary for its development.

In view of the inevitable systematic generation of residues, this work proposes a solidification processes using residues as SSA, specifically the SAP/fluff pulp (polymer), sanding powder and clay soil. These materials have a low cost, great availability, simple operational process, properties that give them a good performance for this application, the fact that through their use as solidifying agent, added value is provided to products that otherwise would be considered as a waste. Moreover, it was investigated the influence of the amount of each solidifying agent in three variables: on the final moisture of the residue, on the volume occupied by the residue after the treatment, and on the total operational cost. It was performed an optimization to find an operational condition that minimizes the cost and final moisture content less 20% on wet basis (w.b.).

2. Methodology

2.1 Materials

The waste management company based in Uberaba – Minas Gerais, Brazil, supplied the materials used in this work. Sanding powder is a waste generated by a company that manufactures wood panels. The SAP/Fluff is a waste generated during the production of personal care products, as diapers. Both residues, SAP/Fluff and sanding powder, are bought by the waste management company at a low cost from another companies. The clay soil used is abundant material in the company, due to the opening of the ditches for waste disposal.

The residue used was classified as a hazardous waste, according to Brazilian law ABNT 10.004 (ABNT, 2004). It was composed by a mixture of liquid effluents containing oil, settleable solids, detergents and septic tank residue. This type of waste is collected in several establishments in Uberaba, Brazil, and forwarded to the waste management company.

In order to evaluate the moisture content of the residue after S/S, the samples were disposed in Petri dishes and to submitted to an oven at 105ºC (± 5ºC), After that, samples were placed in a desiccator, until the weighing, using an analytical balance (precision ± 0.001), and packed in calibrated plastic flasks.

2.2. *Experimental Design*

In order to optimize the S/S, an orthogonal central composite design (CCD) was performed to evaluate the influence of the amount of each solidifying agent (clay soil, sanding powder, and SAP/fluff) on the responses (Y): cost, compaction (volume of compacted waste) and final moisture. Table 1 shows the real and coded variables.

Table 1: Coded variables.

Material	**.-α**	**.-1**	**0**	**1**	**α**
X_1 – Clay Soil [g]	0.4625	29.5000	80.0000	130.5000	159.5375
X_2 – Sanding powder [g]	0.0625	1.5000	4.0000	6.5000	7.9375
X_3 - SAP/fluff [g]	0.0900	2.8000	4.5000	7.3000	8.9100

2.1. Compaction Test

The tests to verify the residue final compaction were carried out in a properly calibrated plastic bottle of 20 ml. After mixing of all materials, according the CCD, each solidified sample was placed in a flask and compacted until the prefixed volume. The compaction was made manually. Then, the residue mass was weighed. The compaction coefficient (*CC*) was calculated by equation (1):

$$CC = \frac{m}{V} \quad (1)$$

where CC is the compaction coefficient; m: mass of solidified residue; *V* is volume occupied by the residue (fixed in 20 mL). From the CC found it was possible to estimate the final volume occupied by all solidified residue in each experiment through the equation 2:

$$V_f = \frac{m_t}{CC} \quad (2)$$

where V_f is volume occupied by the residue after treatment and compaction and m_t is the total mass.

2.2. Residue Final Moisture

The moisture content expresses the amount of water in the residue after S/S. Aliquots were drawn from each experiment, weighed and dried at 105°C (± 5ºC) during 24 hours. After that, the sample was placed in the desiccator until room temperature and weighed. The residue final moisture was obtained from equation (3):

$$U' = 100\frac{M_a}{m_t} \tag{3}$$

where U' is the moisture content on the wet basis (% w.b.); M_a is mass of water and m_t is total mass of the moist material.

2.3. Cost Analysis of the S/S

In order to evaluate the total cost of the S/S based on the amount of SSA used, a cost function for the operation was established. In the equation are considered expenses with equipment, fuel, labor, administrative expenses, cost of solidifying agents, and outsourced services. Investment costs were not considered.

Aiming to calculate the cost of obtaining the clay soil, the cost of excavation and transportation was taken into account, since this material is obtained in the company. According to the reality of the company, that receives not just hazardous waste, the following considerations were made to obtain the cost function [$/ton of effluent treated] presented in Equation (4):

$$Cost = D + DT + B + L + AD + ST + OS + P_P X_P + P_{CS} X_{CS} + P_{SP} X_{SP} \tag{4}$$

where D is the diesel cost; P_P is the price per ton of polymer; X_P is the percentage of polymer used; P_{CS} means the price of the ton of clay soil; X_{CS} is the percentage of clay soil used; P_{SP} is the price per ton of sanding powder; X_{SP} is the percentage of sanding powder used. The dump truck cost (DT) takes into account that 75% of the diesel consumed by the truck is in the S/S process.

Regarding to the backhoe cost (B), it spends 80% of the time on S/S and about 80% of the diesel used by the backhoe is in the S/S. About the labor cost (L), 50% of the work time of the technician responsible for the Class I landfill is engaged in activities involving the receipt and disposal of liquid residues; about 80% of the working time of the Class I landfill helper is engaged in the solidification operation.

The administrative costs (*AD*) was calculated per ton of waste and another outsource services cost (*OS*) were considered, once 50% of the costs of outsourced services were attributed to Class I liquid wastes.

3. Results and Discussion

In this work, the use of different materials in the solidification process was studied in order to determine the appropriate proportion of material to solidify/stabilize a hazardous waste, with a lower cost. Table 2 summarizes the results found for the volume occupied by the solidified residue (V) in disposal, the final moisture (*U'*) and the total cost of the operation for each condition proposed in the experimental design.

Table 2: Experimental planning and responses obtained in S/S of 20 cm^3 of residue: final volume, humidity, and cost of the operation (X_1: Clay Soil; X_2: Sanding powder; and X_3: SAP/fluff).

	Coded Variables			Responses Evaluated		
	X_1 (Clay Soil)	**X_2** (Sanding powder)	**X_3** (SAP/fluff)	**V [cm^3]**	***U'***	**Cost [$/ton effluent]**
Exp. 1	-1.00	-1.00	-1.00	44.36	0.80	48.11
Exp. 2	-1.00	-1.00	1.00	52.01	0.67	53.41
Exp. 3	-1.00	1.00	-1.00	53.87	0.65	48.70
Exp. 4	-1.00	1.00	1.00	85.34	0.56	54.00
Exp. 5	1.00	-1.00	-1.00	135.37	0.33	50.97
Exp. 6	1.00	-1.00	1.00	159.23	0.08	56.27
Exp. 7	1.00	1.00	-1.00	167.74	0.10	51.56
Exp. 8	1.00	1.00	1.00	182.20	0.10	56.86
Exp. 9	-1.58	0.00	0.00	33.39	2.69	49.58
Exp. 10	1.58	0.00	0.00	205.25	0.32	54.09
Exp. 11	0.00	-1.58	0.00	84.11	0.39	51.38
Exp. 12	0.00	1.58	0.00	114.43	0.36	52.30
Exp. 13	0.00	0.00	-1.58	83.01	0.36	46.65
Exp. 14	0.00	0.00	1.58	122.31	0.30	57.02
Exp. 15	0.00	0.00	0.00	98.81	0.34	51.84
Exp. 16	0.00	0.00	0.00	96.61	0.32	51.84
Exp. 17	0.00	0.00	0.00	98.66	0.40	51.84
Exp. 18	0.00	0.00	0.00	97.47	0.30	51.84
Exp. 19	0.00	0.00	0.00	105.61	0.40	51.84
Exp. 20	0.00	0.00	0.00	98.44	0.43	51.84
Exp. 21	0.00	0.00	0.00	106.19	0.34	51.84

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3.1. Effect of solidifiers amount on the packed waste after S/S

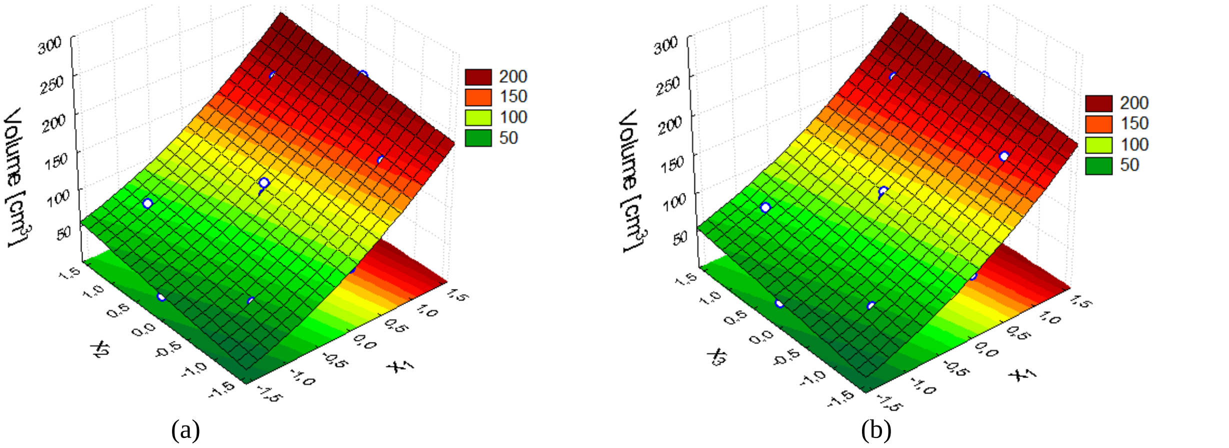
Table 3 shows the results obtained by the solidification process containing only the terms that significantly influenced the final volume of the residue after S/S. All variables in the isolated form had influence on the final volume. It can be noted that the clay soil is the factor that has the greatest influence on the result obtained. Besides, in relation to its isolated effect there is also a quadratic term. Through the analysis of the effect of the isolated variables, it can be noted that the amount of clay soil has an influence about 5 times greater than the amount of sanding powder and the SAP/fluff.

Figure 1 refers to the response surfaces for the residue final volume after S/S obtained from the values presented in Table 2. The increase in the amount of clay soil and sanding powder leads to an increase in the final volume as shown in Figure 1a. The same can be said for the final volume taking into account the amount of clay soil and SAP/fluff used (Figure 1b).

Table 3: Effect of the variables in the residue final volume after S/S [cm³], for treatment of 20 cm³ of effluent (R^2 = 0.9903).

Factor	Effect	Deviation	Significance Level
Average	100.9499	1.3769	0.0000
X_1	104.8723	2.7288	0.0000
X_1^2	16.1170	2.8002	0.0000
X_2	22.5187	2.7288	0.0000
X_3	21.4991	2.7288	0.0000

Figure 1: Response surface for the residue final volume after S/S as a function of the amount of: (a) clay soil (X_1) and sanding powder (X_2); (b) clay soil (X_1) and polymer (X_3) used.



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The volume occupied by the packed waste after solidification can be represented by equation (5). The coefficient of determination was 0.9903, which means that 99.03% of the final volume of the residue can be represented by the model.

$$V[cm^3] = 100.950 + 52.436X_1 + 8.059X_1^2 + 11.2594X_2 + 10.749X_3 \quad (5)$$

3.2. Effect of Solidifiers on moisture after S/S

Table 4 exhibits the results achieved through regression analysis considering only the terms that had a significant influence on the residue final moisture after S/S process. Only the variables X_1 and X_3 influenced significantly the result. Beyond the effect of each isolated variables (X_1 and X_3), the interaction between these variables was relevant too. There is also a quadratic term of the variable X_1. The residue final moisture decreases with increasing amount of clay soil and SAP/fluff used in the mixture. The result is even more sensitive to variations in X_1, which means that the amount of clay soil used will be a determining factor for reaching the desired moisture.

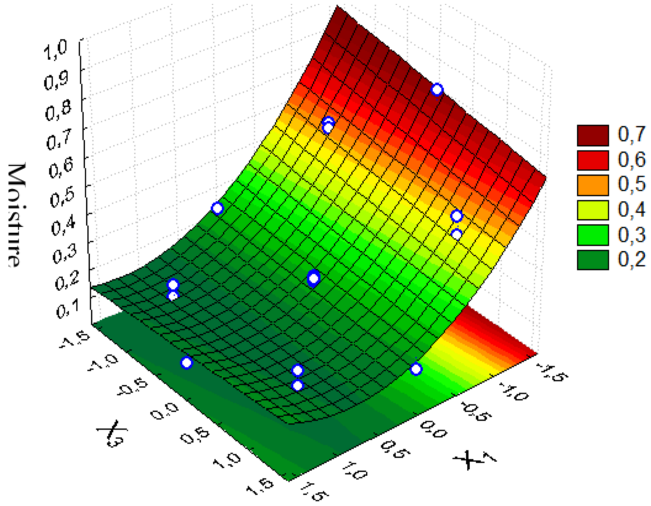
Table 4: Effect of the variables in the residue final moisture (R^2=0.9784)

Factor	Effect	Deviation	Significance Level
Average	0.2200	0.0069	0.0000
X_1	-0.3289	0.0137	0.0000
X_1^2	0.1672	0.0141	0.0000
X_3	-0.0239	0.0137	0.1006
X_1X_3	0.0396	0.0175	0.0377

Figure 2 represents the response surface for the residue final moisture after S/S and it was generated from the values of Table 2. Regardless of the quantity of polymer used, the final moisture content of the treated waste will increase with the decrease of the amount of clay used. In addition, through Figure 2 it is possible to prove the strong influence that the amount of clay soil used will exert on the final moisture. The residue final moisture after S/S can be represented by equation (6) (R^2=0.9784). Regression residuals were random and normally distributed.

$$U_{bu} = 0.2200 - 0.1644X_1 + 0.0836X_1^2 - 0.0119X_3 + 0.0198X_1X_3 \quad (6)$$

Figure 2: Final moisture *(U')* as a function of the amount of soil (X_1) and polymer (X_3).



3.3. Effect of solidifiers on the Cost of S/S operation

Table 5 contains the results of regression, considering only the terms that significantly influenced the cost of the S/S. Only the effects of the isolated variables were significant in the total cost of the S/S. As expected, the amount of SAP/fluff used (X_3) has a greater influence on cost, since this the most expensive SSA. The total cost of the S/S is represented by Equation (7), with R^2= 0.9760. Regression residuals were random and normally distributed.

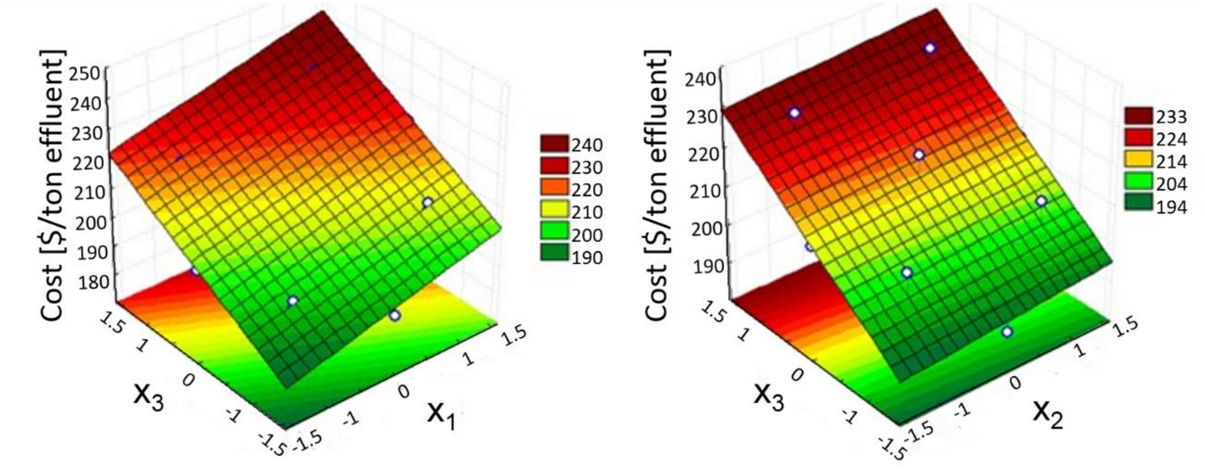
$$Cost\left[\frac{\text{R\$}}{\text{ton of waste}}\right] = 212.5026 + 5.8377X_1 + 1.2000X_2 + 11.8104X_3 \quad (7)$$

Table 5: Effect of variables under S/S cost [R$/ton of effluent], with a R^2=0.9760.

Factor	Effect	Deviation	Significance Level
Average	212.5026	0.3955	0.0000
X_1	11.6755	1.0069	0.0000
X_2	2.4000	1.0069	0.0291
X_3	23.6209	1.0069	0.0000

Figure 3 shows the response surfaces for the S/S cost obtained using the data from Table 2. The operational cost will increase proportionally to the amount of solidifier used. Comparing the results obtained by the variation of X_1 and X_3, Figure 3a, it can be noted that S/S cost is more sensitive to variations in the amount of SAP/fluff than of clay soil used. Through Figure 3b it can be seen that the influence of the amount of SAP/fluff is even stronger when compared to the amount of sanding powder used.

Figure 3: Response surface for the S/S cost as a function of: (a) amount of soil (X_1) and polymer (X_3) used; (b) sanding powder (X_2) and polymer (X_3).



(a) (b)

3.4. Optimization of the S/S operation

The optimization for S/S aimed to perform the operation with the lowest possible cost and final moisture content of the treated residue less or equal to 20% on wet basis. Through the code created in MatLab, a minimum cost of R$ 192.89/ton of waste was obtained. The minimum cost is obtained using 71.70 tons of clay soil/ton of residue, without the use of the other solidifiers. In order to minimize the cost of the operation, it is appropriated to use that only clay soil in S/S, since the use of polymer and sanding powder did not show significant influence on the desired result for the volume and final moisture.

4. Conclusion

The amount of clay soil used as SSA is the variable that has the greatest influence on the final volume occupied by the residue. The amount of sanding powder and SAP/fluff used in the solidification had also an influence on the result, but their effect is not significant when compared to the effect of the amount of clay soil. The final volume of the residue enhances with increasing amount of SSA.

The residue final moisture is influenced by the amount of clay soil and polymer. The moisture increases with the reduction of the amount of soil and polymer used in the blend. The result is even more sensitive to variations in the quantity of clay soil. In another words, the amount of clay soil used will be determinant to reach the desired moisture. Besides the effect of the isolated variables, there is an interaction between them.

The final cost of the operation will increase proportionally to the amount of SSA used. As expected the amount of polymer, the most expensive SSA, used in the solidification will have a greater influence on the cost. The response surfaces obtained for this variable allow saying that the influence of the amount of polymer is even stronger on the cost when compared to the amount of sanding powder used.

The minimum cost for the operation was R$ 192.89/ton of liquid waste. This value can be reached using 71.69 tons of clay soil for each ton of untreated residue. Finally, for a minimum operational cost, only clay soil should be used as a SSA since the use of polymer and sanding powder did not show a relevant influence on the desired volume and final moisture content. The result achieved for optimization takes into account only the volume and the final moisture of the residue. Other factors such as immobilization of dangerous particles in the solid due to the absorbent and adsorbent properties of these SSA or cases where initial soil moisture is different from zero should be also considered in future studies.

It should be noted that the optimization took into account only the volume and the final moisture of the residue. However, the use of the polymer and sanding powder may be advantageous in cases where there is a need for the immobilization of dangerous particles in the solid due to the adsorbent properties of these SSA or even in cases where the initial moisture of the clay soil is different from zero. Situations like these are common in the daily operation of the company and deserve to be studied in depth.

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