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1. INTRODUCTION

The Brazilian phosphate fertilizer is obtained by wet reaction of igneous phosphate rock with concentrated sulphuric acid giving as final product phosphoric acid and dehydrated calcium sulphate (phosphogypsum) (MAZZILLI et al, 2000).

Approximately 69 million t of this by-product have already been produced by the phosphate fertilizer industry in Brazil, with a production of approximately 5.4 million t/year (SAUEIA et al, 2005). All countries that produce phosphate fertilizer by acid wet process are facing the same problem of finding solutions for the safe application of phosphogypsum in order to minimize the impact caused by the disposal of large amounts of this by-product. In Brazil, phosphogypsum (PG) has been used especially in agriculture as soil conditioner. However, the generation rate still much higher than the used one and, therefore, this material is stored in piles close to the site production.

Chemical analysis of air-dried phosphogypsum indicated that, in general, phosphogypsum is enriched with a sulfate compound (CaSO₄) at a level as high as 70%. The sulfate-enriched phosphogypsum can be used in the anaerobic (oxygen depleted) environment, such as landfills, to enhance microbiological processes to decompose municipal solid waste, and thus, extend the lifetime of landfill (SHIEH, 2008).

2. OBJECTIVE

This study aimed to assess, through laboratory studies, whether the application of phosphogypsum as a material for landfill cover in Brazil can contribute to an increment to human dose due to the exposure to natural radionuclides.

3. MATERIALS AND METHODS

The study was carried out in three stages:

- 1) Sampling and analysis of phosphogypsum and municipal solid waste (MSW) (see Fig. 1). The mixture of waste material included organic matter (70%), paper (19.2%), plastics (6.7%), textiles (1.5%), wood (1.5%) and others (1.1%).
- 2) Determination of the phosphogypsum solubility in water according to the Brazilian legislation ABNT NBR 10006 and 10004 (2010).
- 3) Assembly of an anaerobic biodegradation system (vide FIG. 2). In this case, experiments were carried out to measure the reduction in volatile solids in a period of approximately one (01) year and, consequently, to determine the degree of biodegradation of MSW. Four (04) sets of experiments were carried out with the following PG:MSW ratios: 5:0, 5:1, 3:1 e 2:1, which were identified as T1, T2, T3 e T4, respectively. All the experiments were performed in triplicates.

- 4) Analysis of natural radionuclides concentration present in leachate samples by gamma spectrometry (²²⁶Ra, ²¹⁰Pb) and neutron activation analysis (²³⁸U and ²³²Th).



FIG. 1. Segregation and collection of municipal solid waste (MSW).



FIG. 2. Anaerobic reactors with different fractions of PG and MSW.

4. RESULTS

The solid waste solubility test showed that PG was considered neither corrosive nor reagent. The material was classified as noninert waste, based on NBR 10004/2004 (Class II A, non-dangerous/non-inert) due to the presence of As, F, Al, Fe, Mn, and sulfates above threshold limits in the dissolution/solubilization test.

Table 1. Results of pH, humidity and total volatile solid.

Experiment	pH	Humidity	Total Volatile Solid
Before the start of the experiments			
T1	6.6 ± 0.3	58.7	87.6 ± 0.3
T2	6.2 ± 0.9	47.0	47.1 ± 1.2
T3	6.0 ± 0.2	40.0	69.1 ± 12.6
T4	5.5 ± 0.5	39.3	52.9 ± 2.7
After the end of the experiments			
T1	5.6 ± 0.2	60	86.5 ± 0.4
T2	5.4 ± 0.3	60	44.7 ± 8.5
T3	5.7 ± 0.1	60	31.5 ± 2.6
T4	5.8 ± 0.1	60	26.1 ± 0.7

Table 2. Activity concentration of the leachate samples.

Experiment	Radionuclides Activity Concentration (Bq.L ⁻¹)				
	U-238	Th-232	Ra-226	Ra-228	Pb-210
T1	<0.06	<0.03	<0.3	<0.3	2.4 ± 0.7
3	<0.06	<0.03	0.4 ± 0.1	<0.3	0.32 ± 0.03
T3	<0.06	<0.03	<0.3	<0.3	<0.3
T4	<0.06	<0.03	<0.3	0.32 ± 0.09	<0.3
Drinking water limits	10	1	1	0.1	0.1

5. CONCLUSIONS

- As a result of a series of operational testing, an effective equipment configuration for the biodegradation of simulated landfill materials involving application of phosphogypsum was developed.
- The 3:1 ratio of waste/phosphogypsum was demonstrated to be the best applicable condition for landfill application. The radionuclides concentration of the leaching material generated by all set of experiments were below the drinking water limit established by the World Health Organization (WHO, 2008).
- Additional studies are being carried out using a 2D computer model in order to simulate the water flux and radionuclides transport in groundwater due to the application of the ideal ratio of PG/MSW (3:1) in a real landfill. This study will contribute for the evaluation of the radiological risk associated to this practice

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