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Review Article

Phosphogypsum Management Perspectives. Massive Valorization or Massive Storage?

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Abstract

Phosphogypsum (PG) management is a challenge to the authorities worldwide especially because of environmental concerns. PG valorization has been essential as its storage has reached critical quantities. Several studies were carried out in this context to focus on the possibilities of PG valorization especially in agriculture and civil engineering fields as massive valorization axis. However, PG valorization is still quite limited which makes of its storage into embankments a temporary alternative to manage it. Although, PG deposit into embankments is ground intensive and limited by the critical areas and height they can reach. The study of new alternatives is progressing to help optimizing storage areas till putting PG valorization in action.

Keywords: Phosphogypsum; Massive; Valorization; Storage; Environmental; Geotechnical

Abbreviations

PG: Phosphogypsum; CTG: Chemical Tunisian Group.

Introduction

Till 2017, the world counts a population of more than 7 milliards according to the United Nations statistics. World Population Prospects [1]. Providing the nutritional needs of such a population is a challenge that cannot be solved based only on the biological fertilizers. Hence, the use of chemical fertilizers is inevitable with an annual progression ratio of 5% in 2009 according to the MITSI (Moroccan Institute of Technical and scientific information). Phosphogypsum is an industrial residue resulting from phosphoric acid (P_2O_5) which is used in chemical fertilizers production. However, the production of 1tone of P_2O_5 results on 5 tons of PG TCG (2014). Till 2013, the production of Phosphogypsum counts 3 Billion of tones around the world IAEA [2]. This residue used to be either stored in the vicinities of the production factories or rejected into the sea. Nevertheless, these two methods of management showed critical limitations. In fact, PG storage requires intensive areas Bouassida [3], Bouassida [4], Chaari [5], Maazoun and Bouassida [6] while its reject into the sea causes its pollution and threats the maritime fauna and flora IAEA [2]. Hence, Phosphogypsum valorization has been subject of several researches for more than half a decade now. Various valorization options have been considered leading to conclude that agriculture is the more PG consumer method Hilton [7].

Phosphogypsum Management Methods

Phosphogypsum can be managed according to three methods. It is either rejected into the sea, or stored into embankments or valorized.

Phosphogypsum deposit into the sea

Phosphogypsum used to be deposited into the sea, as early experienced in many countries like the United States of America, Spain and the United Kingdom. However, its reject into the sea was forbidden in these countries since the 1990's for environmental concerns while the Moroccan continued depositing more than 15 million tons of PG annually into the Atlantic Ocean IAEA [2], International Maritime Organization [8]. Nevertheless, although Tunisia still rejects PG in Gabes Gulf, of the Mediterranean Sea, it is considered as a world leader in the PG contaminated areas remediation via TAPARURA project, occurred to repair the environmental alteration caused to Sfax City coast by the NPK factory IAEA [2].

Phosphogypsum storage into embankments

Among the 3 billion tons of PG produced till 2013 IAEA [2], 85% are either rejected into the sea or stored into embankments in the vicinities of phosphoric acid production factories Moalla., *et al*. [9]. This method of management allows storing important quantities of Phosphogypsum in specific areas. However, many embankments around the world have known considerable extensions such as the wet deposited embankment of Sfax, Tunisia, with 56m of height, 53Ha of area and 32° of slope Bouassida [4] and the dry deposited embankment of Skhira city, Tunisia too, 55m height, 112Ha area and 1/4 to 2/3 slopes Chaari [5]. The wet phosphogypsum embankments in Huelva, Spain, does not exceed 28m of height Valverde-Palacios., *et al*. [10], this of Mianzhu City, Baiyi Village, China, is almost of 20m height www.greenpeace.org [11], while the New Wales facility wet embankment at Mulberry, Florida, is expected to reach almost 91m in height by 2023 [12].

Phosphogypsum valorization

The United States Environmental Protection Agency (USEPA) classified Phosphogypsum as a Technologically Enhanced Naturally Occurring Radioactive Material (TENORM) Tayibi., *et al*. [13]. Hence, it is rather classified as a co-product, in place of waste. Based on its presence with abundant quantities and its encouraging economic potential, several studies were carried out to focus on the possibilities of its valorization in different fields in an attempt to its use in different ways, especially chemical industry and civil engineering Tayibi., *et al*. [13]. However, the most successful valorization axis ever found is in fertilizing the saline soils, as experienced in Huelva, Spain Valverde-Palacios., *et al*. [10], Hilton [7]. In fact, Phosphogypsum was used for agricultural uses in many countries around the world such as Spain, Australia, Brazil, Egypt, India, Kazakhistan, Pakistan, Syria and the USA. In Brazil, 40% of the produced PG is used for agricultural applications IAEA [2]. In California, USA, an embankment coming from Idaho and Wyoming phosphate was completely used for agriculture before this application was forbidden by the regulation in 1989. In Florida, USA, a small quantity of PG continued to be used in peanut growing although the used PG ratio does not exceed 0.03% of the total PG produced in Florida. In the European Union, PG was used for soil fertilization under the calcium phosphate category IAEA [2].

Soil fertilizing with Phosphogypsum

The soils fertilization using Phosphogypsum allows land repairing (rehabilitation). It recovers the soil productivity, prevents crusts formation and improves water retention as well as it ameliorates trees' growing IAEA [2]. Phosphogypsum improves lands, and allows recovering saline soils as well. This is achieved by reducing the soil toxicity by sodium and aluminum, increasing calcium and sulfurs dissolved from PG, and increasing ammoniac and water retention by the soil IAEA [2]. The use of Phosphogypsum as a fertilizer, generally at a rate of 100-600 kg/Ha, proved its efficiency for many species like carrots, lemons, peach, sugar, avocado, coffee, pepper, beans, corns, tea, cotton, rice, tomatoes, beetroots, onions, peanuts and oranges IAEA [2].

Phosphogypsum valorization in agriculture compared to other fields

In contrast to numerous applications in civil engineering Felfoul., *et al*. [14], Tayibi., *et al*. [13] and chemical industries Tayibi., *et al*. [13], Colombel [15], Al-Jabbari., *et al*. [16], Singh., *et al*. [17], PG is generally used in its generation state without any modification. Recently produced, wet and unaltered, it is diffused on the land according to conventional diffusion methods. However, it is sometimes required purifying PG from acid water and impurities. Hence, to keep it competitive to natural gypsum which is abundant in many countries, it should be taken into consideration that PG purification should be carried out at the lowest cost. I his context, PG extracted from an embankment from north of Florida, USA, was first exposed to rain water for one year, reducing its pH to 5, before it was exploited in agriculture IAEA [2]. Although PG valorization in road field promises the mobilization of big quantities of Phosphogypsum, it is limited by the climate conditions and its application is restricted to arid areas Felfoul., *et al* [14]. In contrast, PG valorization in agriculture is intensive PG consumer. In fact, a big area nearly Huelva, Spain, was recovered using PG and is now among the productive lands (Figure 1) IAEA [2], Hilton [7]. This site is particularly important for the risks evaluation as it is the only referenced site in the world which can be used to study the impact of 40 years of non-stop discharge of PG by land recovery IAEA [2].

This reveals many thoughts to some countries like Tunisia, where almost 1,5 Million Ha of land suffer from soil salinity, which is about 10% of the country's total area Hachicha [18]. Salinity presents a constraining factor for agriculture as the important rate of salts imported by irrigation water can cause lands sterility due to the lack of adequate managements. Hence, the integration of PG in agricultural applications in Tunisia is strongly recommended. It is a valorization option to absorb important quantities of PG, however being lesser than produced quantities.

Figure 1: Land recovered using PG in South West of Spain, Sevilla University IAEA (2013), Hilton (2010).

Limitations of phosphogypsum valorization – need of massive storage

Although several researches revealed that Phosphogypsum can be valorized in civil engineering, chemical industry and agriculture fields, only 15% of the worldwide produced quantity is put into valorization, the other 85% is stored at embankments using either wet or dry process [9]. This is explained especially by the high cost of PG purification which can go through different steps. In fact, PG can be chemically treated by mixing it with ammonium hydroxide aqueous solutions (5-20%) for 24h at 35°C. It is then filtered and washed respectively with ammonium hydroxide solution (0.5%) and water before it gets dried at 42°C temperature which reduces all impurities contents Singh., *et al* [17]. It can be also mixed with citric acid of 3-4% of concentration Tayibi., *et al*. [13]. Several researchers indicated that PG purification can be achieved through many successive wash cycles using demineralized water Felfoul., *et al*. [14], eventually joined to its calcinations Al-Jabbari., *et al*. [16] or neutralization using calcium hydroxide Tayibi., *et al*. [13]. PG thermal treatment reaching 1000°C gave also good results Singh et Garg [17].

Hence, as PG purification procedures are relatively long based on thermal and/or chemical treatments, valorization potentials are quite limited especially in Tunisia which has the second/forth natural field worldwide. The management of the produced PG which can increase by 12 Million tons annually is a big challenge to the concerned authorities. Tunisia cannot be evolved in PG massive valorization, but massive storage, at least in the very short term CTG (2014).

Problems with PG massive storage – Tunisia case study

The deposit of Phosphogypsum into embankments allows storing big quantities of this residue in the vicinity of the production sites. However, these embankments can extend to important areas and height. As the embankments heights and areas increase, several environmental and structural problems appear as a consequence of PG massive storage.

Environmental concerns – Taparura

Phosphogypsum produced by the NPK factory, Tunisia, was deposited in the coast of Sfax resulting in the accumulation of a large square phosphogypsum stack close to the harbour and town (Figure 2.a). The stack, which covered an area of approximately 50 ha and reached a height of up to 8 m above sea level, was surrounded by a crusty layer of phosphogypsum with an area of 90 ha and a depth of up to 3 m IAEA [2]. This caused Sfax coast contamination and a remediation project was launched.

Figure 2: Aerial view of NPK factory deposit site (a) before remediation (b) after remediation.

For Million tons of contaminated materials were removed from the sea and, then, stored on land, to consolidate these residues into a designed central containment. The central containment is a circular terraced structure with a diameter of approximately 0.9 km, an area of 55 ha, a maximum height of 16 m and a 1 in 5 slope (Figure 3). An existing layer of consolidated clay underlies the structure. A layer of uncontaminated soil was used to cap the material contained within the structure (Figure 2.b). The soil thickness varied from 0.8 m (on the top and terraces) to 2 m on the slopes (Figure 3) IAEA [2].

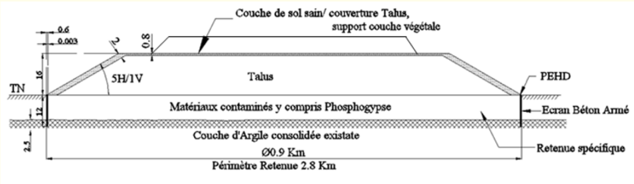

Figure 3: Simplified profile of the remediated PG stack IAEA (2013).

The groundwater level below the remediated stack is constantly monitored by automatic systems IAEA [2].

Geotechnical concerns

In 2012, it was revealed that the wet PG embankment of Sfax City (Table 1) with 56m height, 53Ha area and 32° slope can only be of 70m height maximum Bouassida [4]. This embankment can reach 100m in height if geotextile reinforcement will be operated. This deposition process is well recommended to ensure better interaction between the embankment and the existing ground surface. Using the dry deposited process, the area of the PG embankment of Skhira City (Table 1) covers 112Ha with two elevation levels of 25m and 55m in 2013 Chaari [5].

Deposited embankment	Dry process Bouassida (2007) and Chaari (2013)			Wet process Bouassida (2012)		
Area (Ha)	53			112		
Height (m)	56			elevation: 25m and 55m		
Geometry and shear strength parameters	Height (m)	c (kPa)	*φ (°)*	Height (m)	c (kPa)	*φ (°)*
Layer 1 (upper)	8	90	30	8	10	32
Layer 2	8	97.2	30	48	41.2	32.3
Layer 3	H (*)-16	104.4	30			
	(Chaari, 2013)					
The whole embankment	H	85	31°			
	(Bouassida, 2007)					
Total settlements (m)	4.72 – 4.91			0.86		
Lat. displacement (m)	≈ 0.4			insignificant		
General aspect	More damaging settlements and displacements apart from the cracks and the slope instability			Regular top and slope Better mechanical behavior of the embankment and the ground surface		
H (*): height of deposited embankment at arbitrary elevation						

Table 1: Properties of dry and wet PG embankments.

However, the dry deposited process results in a damaged embankment profile, excessive settlements and lateral displacements. Therefore, a PG embankment of 100m height cannot be achieved. A reinforcement of the embankment by High Density Polyethylene geotextile (HDPE) layers at increments of 4m from 55m elevation allows reaching 130m of height. However, three main factors limit this solution: First, the Tunisian Phosphogypsum pH is found to be of 2.9 Ajam., *et al*. (2009) and about 3, Felfoul., *et al*. [14] which affects the integrity of HDPE layers. Second, over time, there will be an accumulation of HDPE layers partially included into the embankment inferior layer which is assimilated to a stratum and does not require any reinforcement. Third, the study did not take into consideration the observed cracks within the embankment as reported by TCG engineers.

Geotechnical characterization of a heterogeneous and chemically evolutive material is not evident. In fact, the chemical composition of Phosphogypsum depends on the origin of the phosphate ore, the manufacturing process, the efficiency of the plant and the age of the deposit Choura., *et al*. [19], Sahu., *et al*. [20]. These characteristics evolve over time like the soluble P_2O_5 which content increases as the PG gets older due to rain wash for example Felfoul., *et al* [14]. The color of Phospogyosum is dark at recent age Maazoun and Bouassida [6]. Its unit weight evolves as it gets drained and self-weight compacted and its specific unit weight is of 23.1kN/m^3. Its specific color and smell as well as its low specific unit weight reveal the presence of organic matter. The mechanical characteristics of the wet deposited Phosphogypsum over time were studied basing on three specimens: one extracted outlet filter and the others aged 10 and 50 years respectively and superficial Felfoul., *et al*. [14]. The study proved that the best bearing capacity is obtained for the specimen aged 10 (CBR=51% compared to 49% and 5% for SP3 and SP1 respectively) as well as for the best shear strength (c =73 kPa and φ=37°) and a better mechanical performance and behavior to water. In addition, the fresh Phosphogypsum wash allowed improving the compressive strength which reveals that the decrease of acid and organic contents enhances the compressive strength of Phosphogypsum. Hence, the chemical evolution of Phosphogypsum is essentially due to the variation of geotechnical parameters rather than its age Felfoul., *et al*. [14].

In addition to Phosphogypsum mechanical aspect, observations deduced from field visits carried out in March 2017 proved the presence of water retained within the embankment mass Maazoun and Bouassida [6]. At a larger scale, experience with dry storing indicates that the lower layer of the embankment will be saturated even in desert, in arid climate and without rain infiltration due to gypsum self-weight consolidation and settlement Fuleihan [21]. This occurs to some extent in all dry stacks even when Phosphogypsum is well filtered (water\ content << 25%).

New alternatives for phosphogypsum management – recuperation of old/ abandoned embankments

PG storage into embankment can absorb important quantities of this residue instead of rejecting it into the seas. However, PG embankments start to show structural and geotechnical problems as their height reach critical elevations. As PG valorization is quiet limited especially for the PG purification high cost, new alternatives should be found to reduce land use for Pg storage. The recuperation of the existing dry deposited PG embankment of Skhira City, Tunisia, and its reuse as support for a new wet deposited one is an alternative that may help optimizing storage areas. Phosphogypsum time dependent aspect was considered in the numerical simulation carried out. The wet deposit cinematic was also taken into consideration Maazoun and Bouassida [6].

All the materials and methods that are used to complete the study should be mentioned.

Results and Discussion

Results and discussion must illustrate and interpret the reliable results of the study.

Conclusion

Conclusion should reflect and elucidate how the results correspond to the study presented and provide a concise explanation of the allegation of the findings. This paper highlighted the problematic challenges faced for Phosphogypsum management which knew considerable evolution depending essentially on environmental concerns. PG valorization is a promising alternative to solve its storage problems, especially in massive axis such as roads and agriculture. However, Phosphogypsum valorization is still limited to 15% of the worldwide produced quantity because of the high cost of its purification which makes it less competitive to natural gypsum. Till putting PG valorization in action, its storage into embankments is a temporary alternative to manage it. Nevertheless, PG storage is equally limited by the storage areas shortage and the critical heights that may reach the embankments. This evolve that the existing storage areas, either abandoned or still in exploitation, should be recuperated, rehabilitation and reused for PG storage with more intensive quantities. Foundation amelioration can be recommended for this purpose.

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Conflict of Interest

There is no financial interest or any conflict of interest, between the authors and any institution or individuals in regard to the content of this paper.

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