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RESEARCH ARTICLE

# POTENTIALLY UTILIZATIONS OF JORDAN PHOSPHOGYPSUM: A REVIEW

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ABSTRACT

Phosphogypsum is a waste by-product of the phosphate fertilizer industry. The phosphate industry in Jordan has generated over 60 million tons of this waste material. Production continues at the rate of 3 million tons per year. PG, stockpiled at Eshidiya Mines and Aqaba Fertilizer complex site. The toxic heavy metal and radionuclides naturally present in the by-product phosphogypsum and their effect on the human health and environment has urged safe uses in agriculture, soil amendment, cement industry, and building construction.

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## INTRODUCTION

Phospho Gypsum (PG) is a waste by-product of the wet process of phosphoric acid production, which contains variable amounts of some impurities such as naturally occurring radionuclides, fluoride, and heavy metals depending on the source (Rutherford *et al.,* 1994; Al-Hwaiti *et al.,* 2014). About5 tonnes of PG are generated for every tonne of phosphoric acid (H3PO4) manufactured. The operative chemical reaction is generalized as

$$Ca_5F(PO_4)_3 + 5H_2SO_4 + 10H_2O \rightarrow 3H_3PO_4 + 5CaSO_4.2H_2O + HF$$

Worldwide production of PG is over 100–280Mt/a (Mayand Sweeney, 1984). However, only 14% of the worldwide production is reprocessed, 28% is dumped into water bodies, and 58% is being stockpiled and may cause huge environmental problems (Carmichael, 1988). Only 15% of world PG production is recycled as building materials, agricultural fertilizers or soil stabilization amendments and asset controller in the manufacture of Portland cement. The remaining 85% is disposed of without any treatment. This byproduct is usually dumped in large stockpiles exposed to weathering processes, occupying considerable land areas and causing serious environmental damage (chemical and radioactive contamination), particularly in coastal regions.

PG is mainly contains impurities such as residual acids, fluorides, sulphate ions, trace metals (e.g. Cr, Cu, Zn and Cd), radionuclides likes Rn-222, Ra-226, U-238, Th-232, and organic matter as aliphatic compounds of carbonic acids, amines and ketones, adhered to the surface of the gypsum crystals (Rutherford *et al.,* 1996; EPA, 1998; Carbonell-Barrachina *et al.,* 2002). Several studies have been emphasized on the beneficial ways of utilizing PG in order to reduce the disposal problem and the environmental implications. These include uses of PG: (1)agricultural fertilizer or for soil stabilization amendments(Al-Oudat *et al.,* 1998; Enamorado *et al.,* 2009; Abril *et al.,* 2008), (2) cement industry as a setting regulator in place of natural gypsum (Potgieter *et al.,* 2003; Akın and Yesim, 2004), in the gypsum industry to make gypsum plaster (Papastefanou *et al.,* 2006),as mineralizer in the burning Portland cement clinker (Van der Merwe and Strydom, 2004), as raw material in the raw mix of cement (Singh, 2002),and in other binder materials (Taher, 2007). Tayibi *et al.* (2009), reported a preliminary study of a PG stabilization process using a sulfur polymer matrix in order to improve mechanical properties of PG concretes. Results show a new PG concrete product with good mechanical properties and low radionuclide contents, and it can be considered a very suitable and efficient option for PG disposal. This paper reviews the environmental health risk assessment associated with Jordan PG uses and their current and future management and the suggested way to value the high amount of phosphogypsum produced.

**Physico-chemical properties of PG:** PG properties are dependent upon the nature of the phosphate ore, the type of wet process employed, the plant operation efficiency, the disposal method, and the age, location and depth of the landfill or stack where the PG is dumped (Arman and Seals, 1990).Al-Hwaiti *et al.* (2005) studied the XRD characterizations of the phosphogypsum detected gypsum and minor levels of quartz and mica. Fluorides and phosphates were present, as well as compounds of aluminum, magnesium, barium, iron and other elements. SEM examinations confirmed the presence of gypsum and minor quartz. Particle size distributions of Al, P, Ca, S, and Si in coarse, medium, and fine fractions, obtained for the total digestion are shown in Table 1. Silicon is enriched in the coarse size fraction, which might be attributed to the fact that this element is associated with quartz mineral phase as reported by XRD (Al-Hwaiti *et al.,* 2005) and SEM.

**Heavy metals in Phosphogypsum:** Mean heavy metals concentrations (ppm) in phosphogypsumis shown in Table 2. Al-Hwaiti *et al.* (2010a) investigated the particle size distributions of total As, Cd, Cr, Cu, Mn, Mo, Ni, Pb, Se, V, and Zn are shown in Table 3. Zinc was the most abundant heavy metal in both Aqaba and Eshidiyaphosphogypsum. The lowest values observed were for Cd which was slightly less abundant than Cu. Fine particles (0.053 mm) were slightly enriched in most of the trace metals; but no analyze shows clear tendency towards enrichment in small particles

**Radionuclides in phosphogypsum:** Radionucides, as mentioned above PG contains relatively high levels of U-series radionuclides naturally present in the phosphate rock. Depending on the quality of the rock source, PG can contain as much as 60 times the levels normally found prior to processing. The most important source of PG radioactivity is reported to be $^{226}$Ra (Al-Hwaiti *et al.,* 2010b; Al-Hwaiti and Ranville, 2010). Some authors have also reported high $^{238}$U and $^{210}$Po activity. Ra-226 produces radon gas ($^{222}$Rn), which has a short half-life of 3.8 days, an intense radiation capacity, and causes significant damage to internal organs. Particle size distributions of radio nuclides are shown in Tables 3. The distribution was presented by coarse, medium and fine. All analyzed elements are enriched in the fine size fraction. The mean (±SD) of $^{226}$Ra concentrations in ten samples of Jordan PG is 601 ± 98 Bq/kg, which falls near the midrange of values reported for PG samples collected worldwide. Jordan PG generally shows no analytically significant enrichment (>10%) of $^{226}$Ra in the finer (>53 lm) grain size fraction (Zielinski *et al.* 2011;Al-Hwaiti and Ranville, 2010). Phosphogypsum samples collected from two industrial sites with different sources of phosphate rock feedstock show consistent differences in concentration of $^{226}$Ra and rare earth elements, and also consistent trends of enrichment in these elements with increasing age of PG. The results showed that the percentages of $^{226}$Ra and $^{210}$Pb in PG are over those in the corresponding phosphate rocks (PG/PR), where 85% of the 226Ra and 85% of the $^{210}$Pb fractionate to PG. The sequential extraction results exhibited that most of $^{226}$Ra and $^{210}$Pb are bound in the residual phase (non-$CaSO_4$) fraction ranging from 45%–65% and 55%–75%, respectively, whereas only 10%–15% and 10%–20% respectively of these radio nuclides are distributed in the most labile fraction.

**Jordan Phosphogyspum uses**

**Application of PG in Agriculture:** Al- Hwaiti *et al.* (2015d) conducted heavy metal contamination in soils and vegetables (tomatoes and green peppers) and to evaluate the possible health risks associated with the consumption of vegetables grown in PG-amended soils. The enrichment factor values indicated that Pb,Cr, Cu, Ni, Zn, and V were depleted to minimally enriched, and Cd was moderately enriched. The pollution load index values indicated that the PG amended soils were strongly polluted with Cd, moderately polluted with Cr and Ni, and slightly polluted with Pb, Cu, Zn and V. The geo-accumulation index values indicated that the PG-amended soils were uncontaminated with Pb, Cr, Cu, Ni, Zn, V, and moderately contaminated with Cd. The trace metal transfer for Cd, Cr, Pb, and Zn concentrations was below what are considered as acceptable limits (<1)for food production in soil and vegetables (tomatoes and green peppers) at each site area. Soil-to-plant transfer factor values decreased in order of Zn>Pb>Cd>Cr. The biological absorption coefficients in plants are, in order of highest to lowest, Pb>Zn>Cd>Cr, which suggests that Pb is more bioavailable to plants than Cd, Cr, and Zn. Furthermore, this study highlights that both adults and children consuming vegetables (e.g., tomatoes and green peppers) grown in PG-amended soils ingest significant amounts of the metals studied. However, the daily intake of metals (DIM) and the health risk index (HRI) values are >1, indicating a relative absence of health risks associated with the consumption of vegetables/fruits grown in PG-amended soils. However, while DIM and HRI values suggest that the consumption of plants grown in PG-amended soils is nearly free of risks, there are other sources of metal exposures such as dust inhalation, dermal contact, and ingestion (for children) of metal-contaminated soils, which were not included in this study.

**Cement industry:** Al-Hwaiti, M., (2015b) treated Phospho Gypsum (PG) with lime-water (LWT), Sulphuric acid (SAT), a mixture of H2SO4 and HNO3 (AWT), PG-water (ST), and PG-limestone (LT) was attempted to purify PG and improve its quality so that it can be used for manufacture of ordinary Portland Cement (OPC). The treatment of PG removes $P_2O_5$, $SO_3$, and MgO impurities into water-leachable phase. Chemical analysis of the treated PG and mechanical properties of OPC mortar after various treatment of PG established improvement of the quality of PG. The purified PG contain less impurities when compared with untreated PG. It was observed that the leachable of P2O5, SO3, and MgO in these samples ranged from 86% to 90%, 69% to 94%, 96% to 99%, respectively, can be achieved using these treatment processes. The major phases Alite ($C_3S$), Belite ($C_2S$), Aluminatetricalcic ($C_3A$), and Tetra-calcium aluminoferrite ($C_4AF$), and control ratios Lime Saturation factor (LSF), Aluminum/Iron ratio (AR), and Silica ratio (SR) were measured. These experimental results showed that the $C_3S$, $C_3A$ and $C_4AF$, $C_2S$, LSF, AR, and SR contents fulfilled the requirement of the Jordan Standards and European Standards; hence treated PG can be replaced by natural gypsum. The X-ray diffraction analysis of OPC samples showed that $C_3S$ and $C_2S$ are major mineral phases, $C_3A$ and $C_4AF$ represent as minor constituents while the CaO and MgO represent as trace phases. The effect of treated PG on the mechanical properties of OPC mortar was investigated. The OPC produced with purified phosphogypsum were found to have strength properties similar to those produced from mineral gypsum thus

**Table 1. Particle size distributions of major oxides in Aqaba and Eshidiyaphosphogypsum**

| Material | Aqaba | | | | | | Eshidiya | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Age | Al | P | Ca | S | Si | Age | Al | P | Ca | S | Si |
| Composite | 1 yr | 3.39 | 34.2 | 272 | 230 | 10.2 | 1 yr | 10.8 | 63.9 | 266 | 228 | 7.91 |
| Coarse | | 3.56 | 32.3 | 263 | 221 | 9.50 | | 13.0 | 74.2 | 267 | 228 | 11.2 |
| Medium | | 2.77 | 29.6 | 270 | 238 | 9.33 | | 10.3 | 58.9 | 273 | 240 | 8.09 |
| Fine | | 3.61 | 33.1 | 324 | 258 | 7.40 | | 10.7 | 56.3 | 396 | 337 | 11.7 |

**Table 2. Mean heavy metals concentrations (ppm) in phosphogypsum**

| | Mean concentration (ppm) | | | | |
|---|---|---|---|---|---|
| | As | Cd | Cr | Cu | Zn |
| Aqaba | 5 | 3 | 15 | 3 | 29 |
| Eshidiya | 9 | 1 | 8 | 4 | 23 |

**Table 3. Particle size distributions of heavy metals**

| Aqaba | Material | As | Cd | Co | Cr | Cu | Mn | Mo | Ni | Pb | Se | V | Zn |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 Y | Composite | 0.12 | 0.01 | 0.02 | 0.04 | 0.06 | 0.02 | 0.01 | 0.22 | 0.01 | 0.18 | 0.02 | 0.09 |
| | Coarse | 0.10 | 0.01 | 0.02 | 0.04 | 0.08 | 0.03 | 0.01 | 0.22 | 0.01 | 0.15 | 0.05 | 0.04 |
| | Medium | 0.13 | 0.01 | 0.02 | 0.03 | 0.10 | 0.02 | 0.08 | 0.17 | 0.08 | 0.16 | 0.02 | 0.01 |
| | Fine | 0.11 | 0.01 | 0.02 | 0.04 | 0.07 | 0.03 | 0.02 | 0.23 | 0.02 | 0.18 | 0.02 | 0.10 |

| Eshidiya | Material | As | Cd | Co | Cr | Cu | Mn | Mo | Ni | Pb | Se | V | Zn |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1Y | Composite | 0.13 | 0.02 | 0.03 | 0.05 | 0.04 | 0.20 | 0.01 | BDL | BDL | 0.21 | 0.04 | 0.12 |
| | Coarse | 0.14 | 0.02 | 0.03 | 0.07 | 0.08 | 0.25 | 0.01 | BDL | BDL | 0.23 | 0.05 | 0.17 |
| | Medium | 0.12 | 0.02 | 0.03 | 0.05 | 0.07 | 0.19 | 0.01 | BDL | BDL | 0.21 | 0.04 | 0.13 |
| | Fine | 0.13 | 0.02 | 0.03 | 0.05 | 0.03 | 0.20 | 0.01 | BDL | BDL | 0.22 | 0.04 | 0.09 |

**Table 3. Particle size distributions of radionuclides in phosphogypsum**

| Nuclide | $^{234}Th = {}^{238}U$ | $^{226}Ra$ (measured) | $^{214}Pb = {}^{226}Ra$ (measured) | $^{214}Bi = {}^{226}Ra$ | $^{226}Ra$ (avg.) (calculated) | $^{210}Pb$ |
|---|---|---|---|---|---|---|
| Aqaba, 1 year | | | | | | |
| Composite | 35 | 459 | 469 | 475 | 468 | 425 |
| Coarse | 43 | 484 | 489 | 484 | 485 | 414 |
| Medium | 35 | 434 | 449 | 435 | 439 | 415 |
| Fine | 65 | 464 | 454 | 465 | 461 | 415 |
| Eshidiya, 1 year | | | | | | |
| Composite | 70 | 574 | 601 | 582 | 586 | 566 |
| Coarse | 108 | 584 | 631 | 607 | 607 | 542 |
| Medium | 68 | 562 | 554 | 549 | 555 | 566 |
| Fine | 68 | 838 | 786 | 761 | 795 | 607 |

fulfilling Jordan Standards and European Standards. The present study indicates that the use of PG in OPC manufacture can solve the waste disposal problem thus cleaning our environment at one hand, on the other hand it can save natural raw materials i.e gypsum.

**Phosphogypsum as a building material:** Al-Hwaiti, M (2015a) measured the mean values of activity concentration of $^{226}Ra$, $^{232}Th$, and $^{40}K$are in the lower range of typical values reported for phosphogypsum samples collected worldwide. Al-Hwaiti *et al.* (2014)calculated radiological hazard indices such as the radium-equivalent activity($Ra_{eq}$), the gamma index ($I_{\gamma}$) alpha index ($I_{\alpha}$), the absorbed gamma dose rate ($D_{in}$), and the corresponding annual effective dose ($E_{in}$) were assessed for building materials ford wellings.

The results of assessment exhibit that all phosphogypsum samples are higher than the recommended safe limit for building materials for dwellings, except for the radium-equivalent activity ($Ra_{eq}$). Overall assessment, it can be concluded that the possibility of using Eshidiya phosphogypsum in building materials in proportions lower than 100 % will be safe. The mixture of phosphor gypsum with normal gypsum can dilute the concentrations of natural radionuclides allowing the use of the mixed building materials to be safe from a radiological point of view.

**Phosphogypsum treatment:** Al-Hwaiti, M (2015c) treated Phospho Gypsum (PG) in order to reduce activity concentrations of $^{226}Ra$, which was found to exceed that permitted by international regulations. Treatment methods using hybrid water treatment, sulphuric acid treatment, mixed acid ($H_2SO_4$ and $HNO_3$) treatment, household water treatment and calcium carbonate powder treatment were applied. Reduction of $^{226}$Ra content inphosphogypsum by 80–85 % can be achieved using these treatment processes. The radium equivalent activity ($Ra_{eq}$), gamma index ($I_\gamma$), alpha index ($I_\alpha$), absorbed gamma doserate ($D_{in}$), and corresponding annual effective dose ($E_{in}$) were evaluated for public exposure due to the use of treated phosphogypsum in building materials and for other purposes. The calculated values of the ($Ra_{eq}$), ($I_\gamma$), and ($I_\alpha$) forall the treated phosphogypsum samples are significantly below the recommended upper level of unity used as anindex of radiological hazard. The measured mean value of the ($D_{in}$) is about 20 % lower than the population-weighted average value of 84 nGy h-1 for the indoor absorbed doserate. The estimated values of the indoor annual effective dose for all the treated phosphogypsum samples are significantly below the recommended upper level of 1 mSv. The mean value of the ($E_{in}$) is about 70 % lower than up perlevel of 1 mSv.

**Conclusion**

In the overall assessment, it can be concluded that using treated PG in proportions up to 100 % of building materials and other applications will be safe from the radiation protection perspective. Treated PG improves the strength development when used in OPC and hence it can be used in construction industry for preparation of OPC replacing natural gypsum, which is a valuable ingredient of OPC, to achieve economy. Untreated PG, it can be concluded that the possibility of using in building materials in proportions lower than 100 % will be safe. For agriculture purposes, special consideration should be taken because these results reveal that the application of this amendment at normal rates may result in Cd and Pbconcentrations in vegetables above acceptable limits in the long term, with associated consumption risks due to those metals. Therefore, further studies are needed to determine the growth, performance, and mechanisms whereby such plants are able to thrive in contaminated soils, and metal accumulation of the sespecies in the long term.

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