



Better filterability and reduced radioactivity of phosphogypsum during phosphoric acid production in Morocco using a fly ash waste and pure silica additive

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Abstract

Reactive silica additives, such as clays, can increase the filterability of phosphogypsum (PG) during wet phosphoric acid production from phosphate rock (PR). In this study, the effect of adding inexpensive fly ash waste (34 kg per t PR) together with lower quantities of pure silica (8.5 kg per t PR) on the radioactivity of PG was investigated. The addition of fly ash waste/pure silica reduced the radiological activity of the PG by roughly 30%. The reduction was attributed to decreased activities from ²³⁸U (60% reduction) and ²²⁶Ra (30% reduction) in PG. Besides, P₂O₅ losses were slightly decreased.

Keywords Circular economy · Fly-ash additive · Morocco · Phosphogypsum (PG) · Radiological risk assessment · Wet phosphoric acid (WPA)

Introduction

Morocco is one of the largest phosphate rock (PR) producing countries in the world [1]. Mined PR is further processed to mineral fertilizer using the wet phosphoric acid (WPA)

process. During the WPA process some 5 t phosphogypsum (PG) (dry weight) are produced per 1 t WPA. PR can contain elevated concentrations of naturally occurring radionuclides that transfer in parts to the WPA and the PG [2]. Although the activity of PG is relatively low (0.2–3.0 Bq/g for ²²⁶Ra which is the most relevant radionuclide) it permits utilization of PG as a building material under most national regulations [3, 4] so that some 100–280 million t PG per year are stacked in the vicinity of fertilizer plants worldwide today.

The purification of PG is difficult since the radium (Ra) is chemically very similar to calcium (Ca), so that separation from the PG matrix is complex, usually inefficient, and as a result costly [5, 6]. Economic PG treatment and potential utilization is an active field of research with increased pressure to process PG due to the environmental risks associated with storage and management of this bulk commodity [7–10].

In PR processing the main objective of the WPA industry is to obtain the highest concentration of WPA with maximum yield [11, 12]. The size and shape of the PG crystals is a major factor that effects filterability. If the size of the PG crystals is for instance too small, the resulting low filterability creates undesirable P₂O₅ losses [11]. There are options to modify size and shape of the PG particles. Flocculation agents for instance, produce larger aggregates which can be separated easier from WPA. Increasing PG crystal size is an

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effective strategy to increase WPA efficiency [13]. Different additives can further increase the rate of PG precipitation. These additives include aluminum sulfate, clay, and calcined clay [14], aluminum hydroxide, perlite, active charcoal, and manganese dioxide [15, 16], surfactants [17, 18], phosphonates [19, 20], and active silica [21].

WPA production units in Morocco currently use (relatively) expensive clay as a reactive silica additive. Hakkar et al. [22] recently suggested the use of fly ash instead of clay as a low-cost reactive silica source for WPA production from Khouribga PR in Morocco. This practice would further support valuable rare earths recovery from WPA since it increases the share of rare earths transferring from the PR to the liquid WPA instead of the solid PG [23, 24]. Such a practice may be particularly suitable at the Jorf Lasfar industrial zone in El Jadida, Morocco, where PR is processed to WPA. Furthermore, nearly 50% of the country's electricity is produced at the Jorf Lasfar coal power plant, which produces approximately 500,000 t fly ash and an additional 140,000 t bottom ash per year of which only 30% fly ash is currently utilized in construction [25–28].

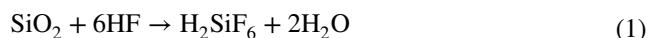
While Hakkar et al. [22] focused on the transfer of rare earth from PR to WPA and PG, this work investigates the possibility of using inexpensive fly ash as a mineral crystal modifier to (1) enhance the filtration rate of PG from WPA and (2) reduce the overall radioactivity of the produced PG so that the PG may find increased use in construction or agriculture in Morocco.

Experimental

Experimental procedure

Experiments were carried out using a laboratory scale unit at conditions simulating the industrial dihydrate process of WPA production in Morocco. The reaction was carried out in a 2 L reactor fitted with a stirrer and placed in a water bath. The phosphate pulp was added continuously using a spatula, while the sulfuric acid (65% H₂SO₄) and the recycled acid (18% P₂O₅) were added drop by drop, using two burettes. All the above feeding was done on continuous bases and the reaction temperature was maintained at 80 °C with constant agitation of 250 rpm. The slurry was agitated for an additional hour to assure sufficient time for PG crystal growth before the PG was filtered. The vacuum filtration was carried out using a Buchner type filter. In the end, the filter cake was washed. These tests were carried out repeatedly until the operating parameters representing industrial conditions that result in WPA with 29% P₂O₅ could be well simulated. All experiments used the same amount of sulfuric acid (250 g per 200 g PR) independent of the use of additives.

A second series of tests were performed in the presence of fly ash, pure silica and a mixture of both materials. The masses of the additives were chosen to maintain a SiO₂/F ratio ≥ 0.53. This ratio was evaluated based on the stoichiometry of the complex forming reaction between F and the equivalent SiO₂ to obtain a total conversion of HF to H₂SiF₆, following (1):



In total, 32 tests (8 baseline experiments without additives, 8 experiments with pure silica, 8 experiments with fly ash and 8 experiments with the fly ash/silica mixture) were carried out to simulate industrial WPA production with and without fly ash/silica additive in Morocco. Mean values of these experiments are presented here.

Chemicals and instrumentation.

Atomic absorption spectroscopy (AAS) was performed using a Perkin Elmer Pinnacle 900 T (Perkin Elmer Inc., Waltham, MA, USA). Elemental analysis was conducted using inductively coupled plasma optical emission spectrometry (ICP-OES) on an iCAP PRO XPS (Thermo Fischer Scientific, Thermo Jarrell Ash IRIS, Waltham, MA, USA) and on an Avio 560 Max (Perkin Elmer, Waltham, MA, USA). X-ray diffraction (XRD) was performed on a Bruker D8 (Bruker Inc., Billerica, MA, USA). The chemicals were sourced from Sigma Aldrich (St. Louis, LA, USA). Analytical grade purity was used if not specified otherwise.

Mineralogical characterization

Silica and active silica were analyzed using AAS. Other major compounds such as Al₂O₃, F, MgO, CaO, and P₂O₅ were determined using ICP-OES. Crystalline phases in PR/PG samples were analyzed using XRD with CuK_α radiation (acceleration voltage of 40 kV and a current of 40 mA). The patterns of diffraction were obtained in a 2θ scanning range from 5 to 70°, with a step of 0.02° and counting time of 11 min. Rietveld method with HighScore software and the current ICSD and PDF4+ databases was used to determine the shares of the mineral phases. The chemical composition of the PR, the fly ash, and the pure silica are provided in Table 1.

The particle distribution of the washed and floated Khouribga PR was further determined using different sieves. More than 85% of the PR showed particle sizes > 40 μm and only 7% of the PR showed particle sizes > 250 μm (Table 2).

Filtration rate

In phosphate industry, the filtration rate is usually expressed in t of P₂O₅ produced per m² per day and can be calculated as shown in (2).

Table 1 Chemical composition of the PR, pure silica, and fly ash

Component	Concentration (wt. %)		
	PR	Pure silica	Fly ash
P ₂ O ₅	30.59	<0.10	<0.10
Al ₂ O ₃	0.43	3.54	5.03
Fe ₂ O ₃	0.22	1.39	3.60
CaO	51.95	46.95	42.20
MgO	0.62	0.65	1.42
K ₂ O	0.08	0.10	0.11
Na ₂ O	0.81	–	–
SiO ₂	2.25	93.00	48.00
F ⁻	4.49	–	–
CO ₂	8.5	–	–

$$FR = \frac{W \times SC \times F}{T} \quad (2)$$

with FR being the filtration rate (t P₂O₅/m²/day), W the suspension weight (g), SC the solids content (%), F the filtration factor = 0.122, T the total filtration, washing and drying time (s). The filtration factor F is related to filter area, conversion of time, weight, and area units, as well as P₂O₅ recovery.

Reaction efficiency, P₂O₅ recovery and washing efficiency

The reaction efficiency is defined as the percentage of P₂O₅ passing from the phosphate concentrate to the WPA. It was calculated as following [29]:

$$\text{Reaction efficiency} = \left(1 - \frac{\%CaO^{PR} \times \%P_2O_5^{PG}}{\%P_2O_5^{PR} \times \%CaO^{PG}} \right) \times 100 \quad (3)$$

$$P_2O_5^{PG} = P_2O_5^{\text{water-soluble}} + P_2O_5^{\text{Unattacked}} + P_2O_5^{\text{Synchrocrystallized}} \quad (4)$$

with: P₂O₅^{PG} being the concentration of P₂O₅ in the PG (%). P₂O₅^{water-soluble}: percentage (%) of water-soluble P₂O₅ in the gypsum cake; CaO^{PR}: % CaO in the PR used to make the acid; P₂O₅^{PR}: percentage (%) P₂O₅ in the PR used to make the acid; CaO^{PG}: percentage (%) CaO in the PG.

P₂O₅ recovery is defined as the percentage of P₂O₅ extracted from the PR into solution, because part of the P₂O₅ is lost if the PG is not fully washed and filtered.

$$P_2O_5 \text{ recovery} = 1 - \left(P_2O_5^{PG} - P_2O_5^{\text{water-soluble}} \right) \frac{\%CaO^{PR}}{\%P_2O_5^{PR} \times \%CaO^{PG}} \quad (5)$$

The washing efficiency is defined as the percentage of water-soluble P₂O₅ that passes from the suspension into the WPA. It is determined as following:

$$\text{Washing efficiency} = 1 - P_2O_5^{\text{water-soluble}} \frac{\%CaO^{PR}}{\%P_2O_5^{PR} \times \%CaO^{PG}} \quad (6)$$

Spectroscopic analysis

The analyses of PR and PG samples were carried out by gamma ray spectroscopy at the National Center for Energy Sciences and Nuclear Techniques (CNESTEN) in Morocco, using a broad energy germanium (BEGe) detector. The PR and PG samples were homogenized, and then packaged in cylindrical bottles with a volume of 175 mL and 75 mL. The energy measurement was 1332.5 keV with a resolution of 1.8 keV. For the energy and efficiency calibration of the BEGe detector, a certified multi energy standard was analyzed under the same conditions and geometry as the samples. The data acquisition, display, and analysis of the amplitude spectrum were carried out using the GENIE 2000 analysis software [30]. These analyses allowed the determination of the specific activity in Bq/kg for each radionuclide present in the samples using the following equation:

$$A(E_\gamma) = \frac{N_{\text{net}}(E_\gamma)}{m \times \epsilon_{E_\gamma} \times t \times I_\gamma \times f_{E_\gamma}} \quad (7)$$

$N_{\text{net}}(E_\gamma)$ represents the net number of strokes of the energy peak E_γ ; m denotes the mass of the sample in kg; ϵ_{E_γ} stands for the counting efficiency for the energy E_γ ; t is the time in s; I_γ is the branching ratio to the energy E_γ ; f_{E_γ} is the correction factor that is dependent on the energy E_γ .

Radiological effects and dose estimation

To evaluate the radiological risks of gamma radiation emitted by radionuclides present in the PR and the PG samples, the radium equivalent activity (Ra_{eq}) was determined. The Ra_{eq} , due to the non-uniform distribution of natural radionuclides in the samples, is generally represented as the sum of the specific activities of ²²⁶Ra, ²³²Th and ⁴⁰K, based on the understanding that 10 Bq/kg of ²²⁶Ra, 7 Bq/kg of ²³²Th and 130 Bq/kg

Table 2 Sieve analysis of the PR

	> 500 μm	> 400 μm	> 315 μm	> 250 μm	> 160 μm	> 125 μm	> 80 μm	> 40 μm
wt. %	0.66	1.35	3.15	6.98	30.08	44.91	67.68	85.79

of ^{40}K would produce an identical gamma dose rate. It is the most widely used index to assess radiological risks. Ra_{eq} was calculated using the following equation [31]

$$Ra_{eq} = A_{226\text{Ra}} + 1,43A_{232\text{Th}} + 0,077A_{40\text{K}} \quad (8)$$

with $A_{226\text{Ra}}$, $A_{232\text{Th}}$ and $A_{40\text{K}}$ being the specific activities in (Bq/kg) of ^{226}Ra , ^{232}Th and ^{40}K in the samples that were analyzed.

Internal and external hazard indices

The hazard indices are defined by a model considering the maximum activity of Ra_{eq} (370 Bq/kg). The external hazard index (H_{ex}) is defined in (9).

$$H_{ex} = \frac{A_{226\text{Ra}}}{370} + \frac{A_{232\text{Th}}}{259} + \frac{A_{40\text{K}}}{4810} \leq 1 \quad (9)$$

In addition to the H_{ex} , the respiratory organs are threatened because of the decay of ^{226}Ra into ^{222}Rn and its daughter products. To account for this, the maximum permissible activity for ^{226}Ra is reduced by half to 185 Bq/kg. This internal hazard index (H_{in}) is defined in (10).

$$H_{in} = \frac{A_{226\text{Ra}}}{185} + \frac{A_{232\text{Th}}}{259} + \frac{A_{40\text{K}}}{4810} \leq 1 \quad (10)$$

H_{ex} and H_{in} should be < 1 or the unity, which is equivalent to an external dose rate of 1.5 mGy y^{-1} absorbed dose rate and annual effective dose.

Absorbed gamma dose rate

The absorbed gamma dose rate \dot{D} (nGy/h) in air at 1 m height is assessed from natural radionuclides originating from ^{226}Ra , ^{232}Th and ^{40}K . The outdoor external dose and indoor external dose rate were determined as depicted in (11) and (12) based on guidelines provided by UNSCEAR [31]:

$$\dot{D}_{out}(\text{nGy/h}) = 0.462A_{226\text{Ra}} + 0.604A_{232\text{Th}} + 0.0417A_{40\text{K}} \quad (11)$$

$$\dot{D}_{in}(\text{nGy/h}) = 0.92A_{226\text{Ra}} + 1.1A_{232\text{Th}} + 0.081A_{40\text{K}} \quad (12)$$

The annual outdoor effective dose rate (\dot{E}_{out}) as well as the annual indoor effective dose rate (\dot{E}_{in}) were calculated as depicted in (13) and (14):

$$\dot{E}_{out}(\text{mSv/y}) = \dot{D}_{out}(\text{nGy/h}) \times 8760(\text{h}) \times 0.2 \times 0.7(\text{Sv/Gy})10^{-6} \quad (13)$$

$$\dot{E}_{in}(\text{mSv/y}) = \dot{D}_{in}(\text{nGy/h}) \times 8760(\text{h}) \times 0.8 \times 0.7(\text{Sv/Gy})10^{-6} \quad (14)$$

These doses consider a conversion coefficient from the absorbed dose in air as well as an indoor and outdoor

occupancy factor to determine the effective dose received by adults. UNSCEAR [31] reports a value of 0.7 Sv/Gy for the conversion coefficient and 0.2 and 0.8 for the outdoor and indoor occupancy factors.

Results and discussion

Effect of the additives on the filtration rate and process efficiency

To avoid losses of P_2O_5 in the PG and to assure a high rate of P_2O_5 extraction, pure silica, fly ash and a mixture thereof was added to PR deficient in silica and alumina so that the crystal size is modified, and filterability is improved. For each experiment, the filtration rate, the P_2O_5 yields, and losses were determined. Table 3 compares the filtration and reaction data obtained when no additive was applied (Baseline) to those observed in the presence additives.

The best filtration rate was obtained with the addition of fly ash and the mixture, for which the improvement was increased by 0.13 and 0.42%, respectively, compared with the baseline. Improving the efficiency of PG filtration is critical to economic utilization of PR. Both reaction efficiency and P_2O_5 recovery are higher with the addition of the additives. The highest efficiency was recorded when the fly ash and the pure silica were mixed. This was attributed to the decrease of crystallized P_2O_5 and unreacted P_2O_5 losses as shown in Fig. 1.

The increase in reaction efficiencies were 1.05, 3.55 and 3.9%; and the increase in P_2O_5 recoveries were 0.5, 2.45 and 2.7% with the addition of pure silica, fly ash, and the mixture, respectively. The obtained washing efficiencies were within the industrial limits of 97.7 to 98.8%.

Effect of the additives on the PG morphology

PG crystals of uniform size are most desirable for filtration and washing. Figure 2 shows the morphology of the PG samples without additives (Fig. 2a), with a pure silica additive (Fig. 2b), with a fly ash additive (Fig. 2c) and with the fly ash/silica additive (Fig. 2d).

Table 3 Results of filterability and P_2O_5 yields

Type of additive	Baseline	Pure silica	Fly ash	Mixture
Filtration rate (t $\text{P}_2\text{O}_5/\text{m}^2/\text{day}$)	6.27	6.37	6.40	6.69
Reaction efficiency (%)	93.05	94.10	96.60	96.95
P_2O_5 recovery (%)	95.45	95.50	97.90	98.15
Washing efficiency (%)	97.70	98.60	98.70	98.80

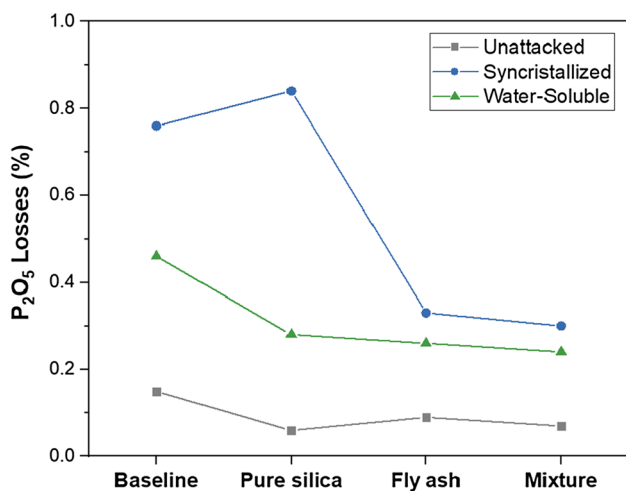


Fig. 1 Comparison of P₂O₅ losses

The microscopic analysis reveals the formation of needle-shaped crystals and small fine diamonds that are characteristic for PG [32, 33]. The increased presence of small crystals decreases filtration by blocking the tissue. The different PG crystal shapes indicate that the additives had a positive effect

on the crystal growth. Specifically, the crystals growth was promoted in one direction leading to fewer small crystals and increased needle-shaped crystals that have a larger length-to-width ratio and can be filtered more easily.

Effect of the additives on the produced WPA quality

WPA prepared from PR has a typical dark green color whose intensity depends on the rock's organic matter content. This coloration is a nuisance, particularly in the preparation of food-grade acid and liquid fertilizers. A quantitative analysis method has also been developed in the laboratory to determine the amount of organic matter contained in the acid. It was based on an analysis by UV/vis spectrophotometer. For a characteristic wavelength of $\lambda = 408\text{nm}$, the absorbance of the strong acid 29% was measured. The results showed a drop of the absorbance from 0.477 to an average of 0.220 in the presence of additives. Doping with additives, thus allowed a clarification of the WPA by decreasing the content of organic matter and fluorine, which opens great possibilities of exploitation such as liquid fertilizers, the production of calcium and ammonium phosphates for animal feed, detergent industry, and surface treatment products. Figure 3b

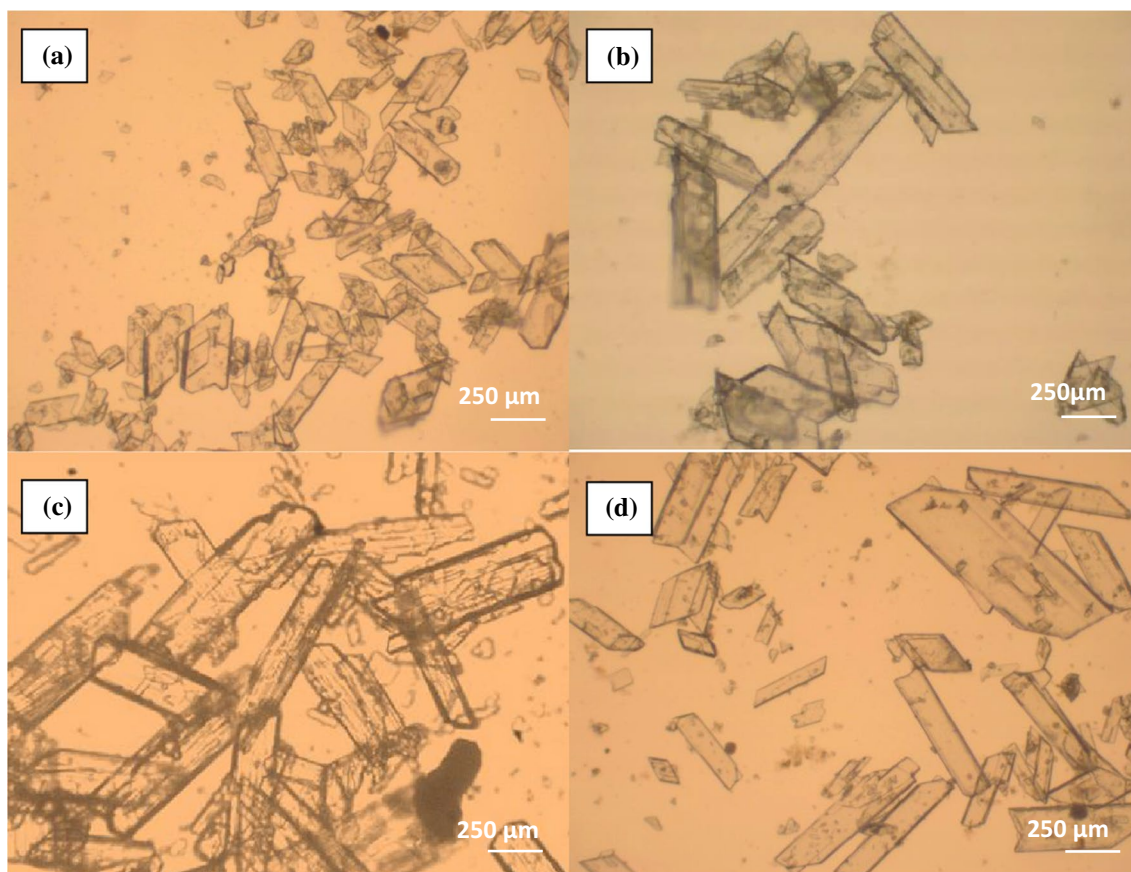


Fig. 2 PG crystals in the presence of: **a** non-additives (baseline), **b** pure silica, **c** fly ash, and the **d** fly ash/silica mixture

shows the produced WPA in its typical dark green and light green color produced with and without fly ash/pure silica additives.

Effect of the additives on the radioactivity of the PG

The specific activities of the three radioactive families (^{238}U , ^{232}Th and ^{235}U) as well as ^{40}K present in the PR and PG samples are shown in Fig. 3a. Most of the radioactivity measured from the PR can be attributed to the presence of radionuclides of the ^{238}U family, particularly ^{226}Ra . The specific activity of ^{238}U was 1,191 Bq/kg and the specific activity for ^{226}Ra was 1,032 Bq/kg. As expected, as PR is a material of natural origin derived from natural geological and precipitation processes, ^{232}Th and ^{235}U families contributed little to the activity of the PR, and the activity of ^{40}K was below the detection limit.

During the production of WPA, the radioactive equilibrium between the three radioactive families and their daughters in the raw PR was broken and each radionuclide was distributed differently depending on its solubility. The results indicate an elevation in the concentration of ^{226}Ra and ^{232}Th in the analyzed PG samples with a percentage of about 85 and 81%, respectively. At the same time, the activities of ^{238}U and ^{235}U in PG is systematically decreased by 81 and 85%. This demonstrates the strong association of the ^{226}Ra , originally existing in the PR, to the PG fraction. The results are consistent with the findings of other studies that had already found that uranium is quite soluble and tends to associate with PA while radium tends to be present in PG [34–37].

The presence of a fraction of ^{238}U detected in the PG was associated with the presence of the remaining P_2O_5 [38,

39]. Whereas radium has been proposed to co-precipitate with $\text{CaSO}_4 \cdot n\text{H}_2\text{O}$ [40]. Radium can be adsorbed by organic material or be mainly associated to BaSO_4 or SrSO_4 solid solutions. The affinity of the radium ions with the barite and strontium crystal lattice can be explained through their similar ionic radius [41]. When the fly ash was added, the fraction of P_2O_5 in the PG decreased and, consequently the activity of associated uranium decreased by 40% from 224 to 89 Bq/kg. The specific activity of ^{226}Ra decreased by 30% from 879 to 617 Bq/kg as shown in Fig. 3a.

The Ra_{eq} value of the PG produced without additives was 897 Bq/kg (Fig. 3b). This value is 2.4 times greater than the current admissible limit for PG use in construction, which is ~ 370 Bq/kg [3, 31]. With the addition of fly ash, the Ra_{eq} value decreases to 641 Bq/kg. It must be concluded that the approach used in this work does not allow for the direct use of the PG as raw material in construction or soil amendment although the radioactivity can be notably reduced.

Radiation hazard indices

Based on the measured specific activity values of ^{226}Ra , ^{232}Th , and ^{40}K in the different samples, the levels of radiological health hazard parameters were calculated, and the results are listed in Table 4.

The calculated values of the H_{ex} and H_{in} for all samples varied from 1.73 to 2.85 and from 3.40 to 5.64, respectively. These values were significantly > 1 , which is the maximum value recommended by UNSCEAR [31]. With the use of the fly ash/pure silica additive, the H_{in} and H_{ex} values could be reduced by 40% due to the reduction of the concentration of ^{226}Ra . Despite the considerable reduction in activity the PG still showed activity levels above the recommended

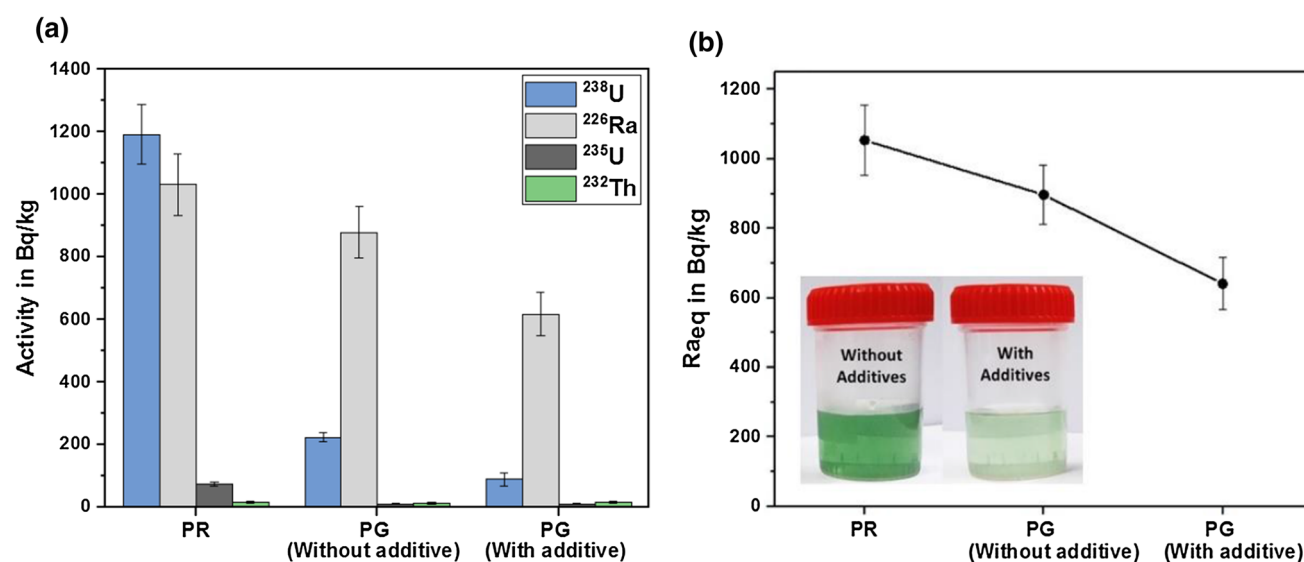


Fig. 3 a Specific activities of the main radionuclides and b Ra_{eq} values obtained for PR and PG samples

Table 4 Radiological hazard indices

Type of sample	H_{ex}	H_{in}	$\dot{D}_{out}(nGy/h)$	$\dot{D}_{in}(nGy/h)$	$\dot{E}_{out}(mSv/y)$	$\dot{E}_{in}(mSv/y)$
PR	2.85	5.64	484 ± 46	967 ± 92	0.59 ± 0.06	4.74 ± 0.45
PG (without additive)	2.43	4.80	412 ± 39	822 ± 77	0.51 ± 0.05	4.04 ± 0.38
PG (with additive)	1.73	3.40	294 ± 34	586 ± 66	0.36 ± 0.04	2.88 ± 0.33

limits so that it cannot directly be used as a raw material in construction.

The measured outdoor (\dot{D}_{out}) and indoor (\dot{D}_{in}) absorbed gamma dose rate ranged from 294 for PG (with additives) to 484 nGy h⁻¹ for PR and from 586 for PG (with additives) to 967 nGy h⁻¹ for PR. These measurements result in outdoor and the indoor annual effective dose of 0.36 to 0.59 mSv/y for \dot{E}_{out} and 2.88 to 4.74 mSv/y for \dot{E}_{in} . Most importantly the values of the outdoor annual dose (\dot{E}_{out}) do not exceed the annual effective dose limit of 1 mSv/y. Not surprisingly, the additives did again reduce the activity of the PG.

By assessing the radiological health hazards associated with PG in the presence of the additives, it can be concluded that even with additives the PG cannot directly be used as a raw material in construction given current regulations. The additives can, however, significantly reduce the activity of the PG so that more of the material could be used in a diluted mixture (if regulations would allow this practice) with natural material while fully complying with the relevant safety standards.

In this context it is noteworthy that sedimentary PR from Morocco and the resulting PG show much higher levels of radiological activity than PR and PG from other regions, particularly igneous PR and resulting PG have considerably lower radiological activity [1]. The findings of this work may thus be most relevant for regions that could reduce the radiological activity of the produced PG by adding inexpensive fly ash/pure silica additives to a point that the PG can be utilized in construction or elsewhere while fully complying to the relevant national regulations. In this study the measured H_{ex} value of PG could for instance be reduced by nearly 30% (28.81%) from 2.43 to 1.73. Qamouche et al. [42] report an average H_{ex} value of 1.48 for Moroccan PG. Assuming a similarly high reduction potential if the fly ash/pure silica additive is used, a H_{ex} value of 1.05 that is very close to the level (< 1) recommended by UNSCEAR [31] could be realized. The same is true for PG and corresponding H_{ex} values reported for Egypt (1.86) [37], the Philippines (1.80) [43] and Serbia (1.64) [44].

Conclusions

The effect of (1) pure silica, (2) fly ash, and (3) a fly ash/pure silica mixture as additives during PR processing to WPA has been investigated. Specifically, the form of the

PG crystals, the rate of filtration and the P₂O₅ yields, as well as the quality of the produced acid and filtered PG was investigated. The results show an increase of the PG filtration rate of 0.13% for fly ash and 0.42% for the fly ash/pure silica mixture. These enhancements of the filtration rate are a result of the modified morphology of the PG crystals that allow slightly better filterability. The reaction efficiency could be increased by 1.05% (pure silica), 2.45% (fly ash) and 3.90% (fly ash/pure silica mixture), and the P₂O₅ recovery was increased by 0.50% (pure silica), 3.55% (fly ash), and 2.70% (fly ash/pure silica mixture). Besides, the use of additives resulted in a cleaner WPA with fewer organic material and reduced fluorine content. The specific activity of ²³⁸U and ²²⁶Ra in PG could also be reduced by 60% and 30%, respectively. Negative effects such as an increased need for sulfuric acid as a result of the additives was not observed.

Although the activity of the PG was significantly reduced using the fly ash/pure silica additives, the PG measured in this study still shows activity levels that do not permit direct utilization under current regulations in Morocco. The results are, however, a promising first step in the right direction and we strongly recommend additional studies that aim at further reducing the activity of PG in WPA production. We recommend that these studies will be conducted with larger material flows to better determine the average radiological activity of the produced PG (that can vary considerably within single stacks) and to better understand if the promising lab-scale results presented here can be reproduced on pilot plant- and subsequently industrial scale.

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