Unconventional Uranium Resources From Phosphates

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Introduction

Phosphate rock or phosphorites are minerals that contain phosphorus (P), measured by its phosphorus pentoxide (P_2O_5) content at elevated concentrations. In phosphate ore the concentrations of P_2O_5 usually range from 5–25%. To be marketed the P_2O_5 is concentrated to roughly 30%. Simple screening and more elaborate techniques such as flotation and calcination are used for the concentration or beneficiation of P_2O_5. P is an essential building block for plants and animals. To feed the world approximately 250 million t of phosphate rock are mined annually. This makes phosphate rock the fourth most mined material on earth. The majority (> 90%) of the mined phosphate rock is used for mineral fertilizer production. As of now there is no substitute for P in fertilizer production and large-scale recycling of P is still in its infancy. Phosphate rock is thus directly tied to food security.

Phosphate rock can contain a number of accompanying valuable elements in elevated concentrations that could justify commercial recovery (depending on specific market conditions and production costs). Most notably, rare earth elements (REE) and uranium could be industrially recovered (**Chen and Graedel, 2015**). Uranium can be found in some phosphate rock in relevant average concentrations of 70–200 mg/kg that could justify recovering it. Uranium is a relatively common element in the earth crust appearing with an average concentration of 2.8 mg/kg. In comparison, the average concentration of uranium in seawater that is, as a result of the tremendous overall quantities of uranium (approximately 4 billion t), often considered as a future uranium resource is as low as 0.003 mg/kg or 3 parts per billion. Uranium resources can be differentiated by concentration (WNA, 2020a,b) into very high-grade ores, high-grade ores, low-grade ores and very low-grade ores as indicated in **Table 1**.

Average uranium concentrations in phosphate rock deposits in Algeria (Djebel Kouif, 100 mg/kg), Angola (Cabinda, 260 mg/kg), Brazil (Araxa, 182 mg/kg and Catalao, 220 mg/kg), Burkina Faso (Kodjari, 125 mg/kg), Egypt (Hamrawein, 110 mg/kg, Safaga, 120 mg/kg, and West Mahamid, 100 mg/kg), Israel (Arad, 150 mg/kg), Mali (Tilemsi, 123 mg/kg), Morocco (undifferentiated, 130 mg/kg), Tanzania (Minjingu, 390 mg/kg), United States (Central Florida, 141 mg/kg and Idaho, 107 mg/kg) as reported by **Van Kauwenbergh (1997)** could thus also be classified as very low-grade uranium ores.

Ironically, several phosphate rock producers mine ores with natural uranium concentrations exceeding those of commercial uranium mines. Uranium from phosphate is considered an unconventional resource, since mining phosphate rock for its uranium content alone or recovering uranium as a by-product is presently considered uneconomic. Should uranium prices increase, or technologies allow for more cost-effective recovery, uranium from phosphates could become a conventional uranium resource again.

It is noteworthy that radiotoxic uranium from phosphate rock, not recovered during phosphate fertilizer production will largely transfer to the final fertilizer product and ultimately be distributed on agricultural soils. There is an active scientific discussion regarding the potential harm of uranium brought out with mineral fertilizers on agricultural soils (**Bigalke et al., 2017**; **Liesch et al., 2015**; **Schnug and Haneklaus, 2015**). While there is presently no legal limit for uranium in fertilizers, the German Commission for the Protection of Soils for instance, proposed setting it to 50 mg uranium per kg P_2O_5 (**Kratz et al., 2016**) or 167 mg uranium per kg fertilizer with a P_2O_5 content of wt30%.

Past uranium recovery from phosphates

Phosphate rock is present as apatite, most commonly fluorapatite ($Ca_5(PO_4)_3F$ or $Ca_{10}(PO_4)_6(F,OH)_2$), that is soluble on only very acidic soils as they are found in some tropical areas. To make P accessible to plants the majority of phosphate rock is processed to soluble mineral fertilizers. The most common process for this is the wet phosphoric acid (WPA) process. During WPA production the concentrated ore is digested with either hydrochloric acid (HCl) (Eq. **1**), nitric acid (HNO_3) (Eq. **2**) or sulfuric acid ($H_2SO_4{\cdot}nH_2O$) (Eq. **3**).

$$Ca_{10}(PO_4)_6F_2 + 20\ HCl \rightleftharpoons 6H_3\ PO_4 + 10CaCl_2 + 2HF \tag{1}$$

$$Ca_{10}(PO_4)_6F_2 + 20\ HNO_3 \rightleftharpoons 6H_3PO_4 + 10Ca(NO_3) + 2HF \tag{2}$$

$$Ca_{10}(PO_4)_6F_2 + 10\left(H_2SO_4 \bullet nH_2O\right) \rightleftharpoons 6H_3PO_4 + 10\left(CaSO_4 \bullet nH_2O\right)_s + 2HF \tag{3}$$

Table 1 Uranium resources by concentration.

Classification	*Uranium concentration*	
Very high-grade ore	20%	200,000 mg/kg
High-grade ore	2%	20,000 mg/kg
Low-grade ore	0.1%	1,000 mg/kg
Very low-grade ore	0.01%	100 mg/kg

WNA (World Nuclear Association) (2020a) *Supply of Uranium*. https://www.world-nuclear.org/information-library/nuclear-fuel-cycle/uranium-resources/supply-of-uranium.aspx.

Phosphate rock digestion with sulfuric acid is the most common process, used in about 75% of the roughly 400 fertilizer plants operating worldwide. The use of sulfuric acid is attractive from an industrial viewpoint as calcium is simultaneously precipitated as solid calcium sulfate known as phosphogypsum ($CaSO_4{\cdot}nH_2O$) that can be separated using simple filtration. The use of hydrochloric acid and nitric acid do in contrast form water-soluble calcium nitrate ($Ca(NO_3)$) or calcium chloride ($CaCl_2$) that require more costly purification techniques. Besides, the majority (80–90%) of the radiotoxic radium transfers to the phosphogypsum replacing Ca in the chemical structure if sulfuric acid is used instead of being liberated as is the case with hydrochloric- and nitric acid digestion. Depending on the crystallization temperature, gypsum dihydrate ($CaSO_4{\cdot}2H_2O$), hemihydrate gypsum ($CaSO_4{\cdot}0.5H_2O$) and gypsum anhydrite ($CaSO_4$) that are usually all referred to as phosphogypsum are produced. For every 1 t phosphoric acid approximately 5 t phosphogypsum (dry weight) are produced. As a result of its low radioactivity most of the produced phosphogypsum (100–280 million t/year) is stacked next to fertilizer plants and not further processed (**Saadaoui et al., 2017**; **Tayibi et al., 2009**). Even the relatively low radioactivity concentrations (0.2–3 Bq/g for ^{226}Ra) present in most phosphogypsum does not permit further using it under most national regulations today. Recent successful case studies on the use of phosphogypsum as construction material and soil amendment are slowly changing the practice of stacking (**Hilton et al., 2020**). Reducing the radioactivity even further by removing radium decay products could make this phosphogypsum available as an inexpensive resource for agriculture and construction. The WPA process for fertilizer production with uranium recovery is schematically shown in **Fig. 1**.

After the phosphogypsum is filtered out uranium can be recovered from phosphoric acid. Phosphoric acid must be purified in the WPA process depending on its further use, as other impurities in addition to uranium such as iron, magnesium, arsenic, aluminum, etc. are dissolved in the digestion stage and transfer into the phosphoric acid. Food and pharmaceutical industries require high-grade phosphoric acid whereas low-grade phosphoric acid is sufficient for fertilizer production.

While the majority (60–90%) of the uranium transfers into the phosphoric acid stream the majority of the REEs will be found in the phosphogypsum, making recovery of the REEs more challenging. For uranium recovery from phosphoric acid, mostly solvent extraction (SX) and to a much lesser extent ion exchange (IX) techniques were used in the past (**Haneklaus et al., 2017a**). Particularly, solvent extraction of uranium from phosphoric acid can be considered an industrially mature technology that has also been reviewed in the scientific literature (**Beltrami et al., 2014**; **Bunus, 2000**; **Singh et al., 2016**) while new ion exchange processes are currently investigated for future uranium recovery from phosphates that will still need to be tested on industrial scale.

Besides, it is also possible to leach uranium directly from phosphate rock using weak acids and conventional leaching techniques (**Al-Khaledi et al., 2019**). Lab-scale experiments that leached uranium and REE from phosphate rock showed that such an approach can be beneficial in a way that uranium is removed early on in the process and at least the radioactivity in phosphogypsum caused by ^{238}U can be dramatically reduced.

Assuming uranium recovery from WPA, the amount of uranium that can be recovered from a certain phosphate rock can be estimated using Eq. **(4)** for phosphate rock and Eq. **(5)** if mass and uranium concentrations of the relevant phosphoric acid are provided.

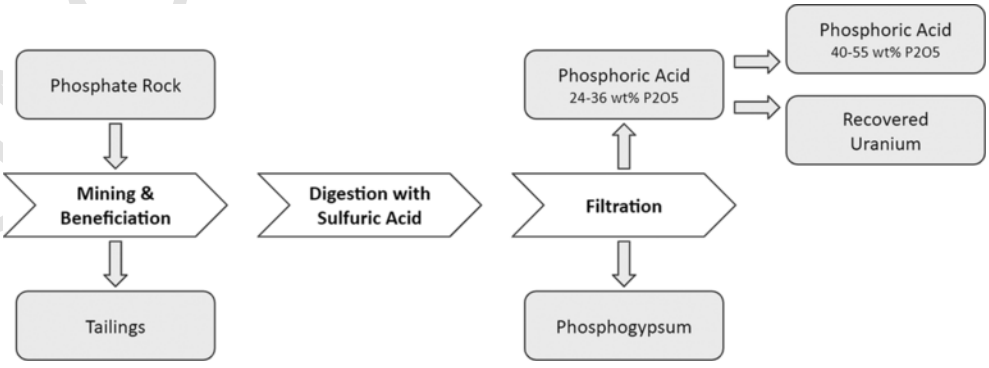

Fig. 1 Schematic overview of the wet phosphoric acid (WPA) process with uranium recovery.

$$m_U = \left(m_{PR} * c_{U,PR} * f_{U,PA}\right) * \eta_{U,PA} \quad (4)$$

$$m_U = \left(m_{PA} * c_{U,PA}\right) * \eta_{U,PA} \quad (5)$$

m_U = Mass uranium, m_{PR} = Mass phosphate rock, m_{PA} = Mass phosphoric acid, $c_{U,PR}$ = Concentration of uranium in phosphate rock, $c_{U,PA}$ = Concentration or uranium in phosphoric acid, $f_{U,PA}$ = Fraction of uranium that transfers from the phosphate rock to the phosphoric acid, $\eta_{U,PA}$ = Extraction efficiency of uranium from phosphoric acid.

The amount of uranium that could thus be recovered from 1 t phosphate rock from Central Florida (m_{PR}) that shows an average uranium concentration ($c_{U,PR}$) of 141 mg/kg (**Van Kauwenbergh, 1997**), if 80% ($f_{U,PA}$) of the uranium transfers to the phosphoric acid and 90% ($\eta_{U,PA}$) can be extracted from the phosphoric acid is a bit more than the mass of a chocolate bar or 102 g.

Larger phosphate fertilizer plants process 500,000 thousand to 2 million t phosphate rock per year resulting in notable potential for uranium recovery. **Table 2** summarizes past commercial uranium recovery activities in the United States, Belgium, Canada, Taiwan and Iraq. Different extractant solvents were used for uranium recovery that gave the respective process its name. Bis(2-ethylhexyl) phosphoric acid (DEPA) and tri-n-octylphosphine oxide (TOPO) was the most popular extractant used for the DEPA-TOPO or ORNL process named after Oak Ridge National Laboratory (ORNL) in the United States where the process was developed.

Uranium recovery activities are sometimes classified in a 1st and 2nd wave of uranium recovery from phosphates (**Haneklaus et al., 2015a**). In the 1st wave (1951–61) uranium mining was still in its infancy and various materials, including phosphate rock, were considered to be potential sources of uranium. With the discovery of higher-grade uranium deposits the 1st wave and uranium recovery from phosphates stopped. The 2nd wave (1976–98) was commercially driven. Since the majority of costs for excavation, mining infrastructure, etc. are covered by the phosphate industry already (**Reitsma et al., 2018**) and uranium prices were favorably high as a result of the oil crisis in 1973, operating commercial uranium extraction plants next to fertilizer plants was economic for some time and done in the United States, Belgium, Canada, Taiwan and Iraq. In addition, uranium was recovered from phosphates in Kazakhstan behind the iron curtain, and many other countries operated pilot plants investigating potential uranium recovery from domestically mined or imported phosphate rock.

Particularly impressive was uranium recovery from phosphates in the United States, where the technology at its height contributed up to 20% of the domestic uranium production in the 1980s before decreasing uranium prices made this process uneconomic. **Fig. 2** compares historic uranium imports and historic uranium mining with historic and potential uranium recovery from phosphates in the United States.

Like many other countries that rely on nuclear power for a considerable share of their electricity production the United States is importing the bulk of the required uranium from abroad. Interestingly, since the 1990s the amount of uranium that could theoretically be recovered from phosphates as a byproduct in fertilizer production could be larger than actual domestic uranium mining. In this context, recent concerns about uranium supply security in the US may indeed *make uranium recovery from phosphates great again* as speculated by **Steiner et al. (2020)**.

Table. 2 Historic uranium recovery from phosphates by country and plant.

Operating period	*Country*	*Plant*	*Uranium extraction process*	*Capacity (t U/year)*
1951–62	United States	Joliet, IL	Precipitation	31
1952–56	United States	Texas City, TX	OPPA	20
1954–59	United States	Nichols, FL	–	–
1955–61	United States	IMCC Plant, Bonnie, FL	OPPA	31
1957–61	United States	Tampa, FL	OPPA	62–138
1976–80	United States	W.R. Grace Plant, Bartow, FL	OPAP	109–127
1978–81	United States	Farmland, Pierce, FL	DEPA-TOPO	153–173
1978–99	United States	Uncle Sam, Convent, LA	DEPA-TOPO	265
1979–82	United States	Riverview Plant, East Tampa, FL	OPPA	138–164
1980–85	United States	CF Industries, Bartow, FL	DEPA-TOPO	231–243
1980–92	United States	CF Industries, Plant City, FL	DEPA-TOPO	231–243
1980–92	United States	IMC, New Wales, FL	DEPA-TOPO	265–288
1980–98	Belgium	Engis, Liège	DEPA-TOPO	50
1980–87	Canada	WCFL, Calgary, AB	OPAP; DEPA-TOPO	38
1981–85	Taiwan	China Phosphate, Lung Tan	DEPA-TOPO	10
1981–98	United States	Sunshine Bridge, Donaldsonville, LA	DEPA-TOPO	162
1984–91	Iraq	Al Qaim	DEPA-TOPO	87

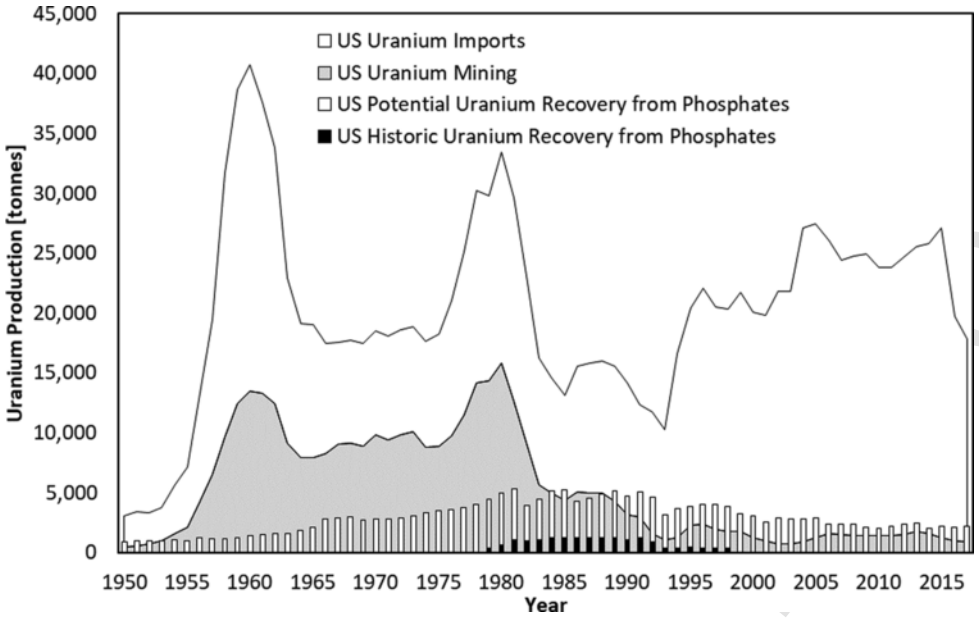

Fig. 2 Historic US uranium imports, uranium mining, past- and potential uranium recovery from phosphates.

Potential uranium recovery from phosphates

The potential of uranium from phosphates can be estimated using annual production data and phosphate rock reserves as they are published annually by the **USGS (2020)** as well as average concentrations of uranium in these phosphates. **Table 3** shows the amounts associated with phosphate rock of the largest 23 producing countries in 2018 as well as the quantity of uranium associated with phosphate rock reserves in these countries. Production data was taken from the **USGS (2020)** and average uranium concentrations by country were taken from **Van Kauwenbergh (1997)**.

The very simple quantitative assessment in **Table 3** indicates that there are roughly 14,000 t uranium associated with phosphate rock production annually. Of these 14,000 t approximately 8,800 t of uranium are associated with phosphate rock that show elevated concentrations of 90 mg uranium per kg that may justify economic recovery. The brief estimate presented here corresponds well to more elaborate studies by Gabriel et al. (2013a,b) and **Ulrich et al. (2014)** that estimate the potential uranium from phosphate rock to be as high as 10,000 t and 11,000 t respectively.

In this context, world uranium production for the same year (2018) was approximately 53,000 t **(WNA, 2020b)**. Equally impressive are the total amounts of uranium associated with phosphate reserves that are with 6.25 million t as high as recoverable identified uranium resources (6.15 million t) (**WNA, 2020b**). Just like the amount of uranium associated with seawater, the quantity of uranium from phosphates is impressive, but will only be relevant if it can be recovered economically.

The economics of phosphate production, the uranium price and environmental benefits of recovering uranium are the most important factors to determine whether uranium recovery from phosphates is economic or not (**Haneklaus et al., 2017b**). In addition, strategic considerations such as uranium supply security may be considered.

To estimate quantities of uranium that may be recovered economically one day a cut-off grade of 90 mg uranium per kg phosphate rock is often used. **Kim et al. (2016)** and **Steiner et al. (2020)** estimated that the quantities of unconventional uranium that could be recovered during phosphate rock processing in the United States could theoretically cover 10% of the national uranium requirements (2,116 t of 20,386 t uranium in 2014). Similar assessments were conducted by **Shang et al. (2021)** for China, indicating that China could have recovered some 648 t uranium during phosphate fertilizer production in 2016. This amount could have covered nearly 10% of the country's reported uranium requirements for that year. The Chinese case study is particularly interesting since the average uranium content in Chinese phosphate rock (27 mg/kg) provided by **Van Kauwenbergh (1997)** in **Table 3** would not justify uranium recovery. More recent investigations by **Ye et al. (2019)** found that there are indeed phosphate rock locations in China with uranium concentrations as high as 480 mg/kg. Even if these deposits are not in the majority, uranium recovery from them should be considered and could add relevant amounts of heavy metal to the country's uranium supply.

The Chinese case study is further relevant as it highlights the importance of relatively small deposits with elevated uranium contents that could actually profit from recovery of most uranium from those phosphates and could be the next locations where this technology may be relevant. Another example of this is the Minjingu phosphate rock deposit in Tanzania that shows elevated average uranium concentrations of 390 mg/kg with peak concentrations reaching nearly 800 mg/kg. The high uranium (and also REE) concentrations of the Minjingu deposit may justify multi-element mining as is already done at Olympic Dam in Australia.

Table. 3 Potential quantities of uranium associated with phosphates.

	Phosphate rock production in 2018 (million t)	*Estimated phosphate rock reserves in (million t)*	*Estimated average uranium content (mg/kg)*	*Uranium associated with phosphate production in 2018 (t)*	*Total amount of uranium associated with phosphate reserves (t)*
Algeria	1.200	2200	63	76	138,600
Australia	2.800	1,200	84	235	100,800
Brazil	5.740	1,700	201	1,154	341,700
China	120.000	3,200	27	3,240	86,400
Egypt	5.000	1,300	90	450	117,000
Finland	0.989	1,000	37	37	37,000
India	1.600	46	23	37	1,058
Israel	3.550	62	120	426	7,440
Jordan	8.020	1,000	84	674	84,000
Kazakhstan	1.300	260	100	130	26,000
Mexico	1.540	30	100	154	3,000
Morocco	34.800	50,000	97	3,376	4,850,000
Peru	3.900	210	72	281	15,120
Russia	14.000	600	28	392	16,800
Saudi Arabia	6.090	1,400	100	609	140,000
Senegal	1.650	50	67	111	3,350
South Africa	2.100	1,400	23	48	32,200
Syria	0.100	1,800	75	8	135,000
Togo	0.800	30	94	75	2,820
Tunisia	3.340	100	44	147	4,400
United States	25.800	1,000	99	2,554	99,000
Uzbekistan	0.900	100	47	42	4,700
Vietnam	3.300	30	30	99	900
Other countries	0.970	770	–	–	–
Total	249.489	69,488		14,353	6,247,288
Total uranium with 90 mg/kg cut-off grade				8,798	5,560,960

Besides the case studies for the United States and China, **Tulsidas et al. (2019)** and **López et al. (2019)** conducted case studies for the European Union where 2% of the uranium requirements or 334 t uranium of 16,084 t total demand could have come from phosphates in 2017 and Argentina where 8% of the uranium requirements or 19 t uranium of 235 t uranium could have come from phosphates in 2017. Both case studies assume a cut-off grade of 90 mg/kg, and in both case studies phosphate rock imports are of significant importance.

All countries need phosphate fertilizer for their food production and several countries have phosphate rock processing facilities without actually mining phosphate rock. Detailed material flows are for instance available for a phosphate rock processing plant in the Philippines **Haneklaus et al. (2015b)** that processes phosphate rock from different parts of the world.

Morocco has by far the largest phosphate rock reserves in the world (73% in **Table 3**) and this phosphate rock shows elevated uranium concentrations of 97 mg/kg that are often reported to actually be as high as 130–160 mg/kg (**Azouazi et al., 2001**). Orano SA (previously Areva SA), the large multinational nuclear fuel cycle company headquartered in France, signed a memorandum of understanding with Morocco's state-owned phosphate rock mining company (OCP Group) in 2006 for uranium recovery from Moroccan phosphates. Unlike the United States and China that are the other major phosphate rock producing countries in the world (see **Table 3**), Morocco does not have a large national market for phosphates and exports most of the mined ore to other countries.

Given the current phosphate rock reserves, Morocco will most likely supply various regions in the world with phosphate rock, phosphoric acid and phosphate fertilizers in the near future (**Cooper et al., 2011**). The increased use of Morocco phosphate rock with elevated uranium concentration could favor uranium recovery. Mineral resources are dynamic in a way that innovations and exploration efforts continuously discover "new" materials deemed economic for mining and processing (**Scholz and Wellmer, 2013**). Seabed phosphates, such as the Chatham Rise deposit offshore of New Zealand with 240 mg/kg uranium content may become a resource soon, changing the current assumptions about resources, reserves and geological potential one more time.

While one day phosphate rock may be mined not only for its P content but for all its constituents such as uranium, REE and its rock matrix that can be used in construction after radiotoxic uranium and its radioactive decay daughters have been recovered (**Zhang, 2014**),

this is clearly not the case yet. The latest push towards commercial uranium recovery from phosphates by PhosEnergy was put on hold in 2019 as a result of low uranium prices. These low uranium prices are presently forcing some commercial uranium mines into a state of permanent maintenance. Facilities to recover uranium from phosphates can be erected fairly quickly (2–3 years) compared to the time required to set-up a new commercial uranium mine (5–10 years). Should uranium prices rise again though, it will most likely be uranium mines currently shut down that will be online first to fill the demand gap.

Conclusions and outlook

Quantities and concentrations of uranium associated with phosphates are considerable, and uranium from phosphates is probably the most important unconventional uranium resource that is not mined today. Although the majority of the costs for excavation, infrastructure development, mining, and processing are covered by the phosphate industry, uranium recovery from phosphates is not profitable today and it is unlikely that this will change as long as uranium prizes are low. However, uranium supply security and environmental concerns may lead to uranium recovery from phosphates at selected locations.

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