

7-1-2014

## Remediation of NORM and TENORM Contaminated Sites—Review Article

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### Recommended Citation

Rahman, R.O. Abdel; Elmesawy, Mohamed; Ashour, I.; and Hung, Yung Tse, "Remediation of NORM and TENORM Contaminated Sites—Review Article" (2014). *Civil and Environmental Engineering Faculty Publications*. 99.  
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# Remediation of NORM and TENORM Contaminated Sites—Review Article

R. O. Abdel Rahman, Mohamed Elmesawy, I. Ashour, and Yung-Tse Hung

## INTRODUCTION

NORM and TENORM are generated in the form of by products, residues and wastes, from large scale industrial processes that exploit natural resources such as uranium mining and milling, fertilizers production, and processing of metal and oil. Table 1 lists activities that lead to generation of NORM and TENORM from mineral ores extraction and industrial processing [1-3]. The management of these materials is receiving considerable attention because of the large volumes of generated NORM, low specific activities, and very long lived radionuclides that exist due to natural radioactive decay of uranium and thorium decay series. In view of these features, NORM/TENORM should be treated as a unique environmental hazard, taking into account the national regulation and regional and international convention.

Historical NORM and TENORM management entailed land spread, surface impoundments and surface mounding. These practices have yielded in several soil, surface water and groundwater contamination problems [1]. It is worthy to note that contamination is defined as the presence of unwanted substance at concentrations above background, while pollution is contamination that results in or can result in adverse biological effects to resident communities [4]. To control these contaminants from spreading leading to greater pollution problems, there is a need to launch a remediation program. Remediation programs are very costly, so the support of the decision making process to initiate such program for contaminated site has received great attention. Usually the decision is made based on two step assessments. At first, the contamination extent is identified by studying radionuclides dispersion pathways and mechanisms in soil surface, aquifers, groundwater, air, and surface water. Subsequently, land use, availability of natural resources in the site and fund availability are assessed. IAEA had defined radiological criteria to support the remediation decision making process, this criteria is based on the definition of six bands for possible remediation situation each cover an order of magnitude in dose or risk [5,6]. Table 2 illustrates IAEA criteria for the beginning of remediation program for contaminated area [5]. The traditional approach to remediate soil contamination was based on removal and confinement, where contaminated groundwater was traditionally remediated using treat and pump technique [7]. Due to strengthen regulatory requirements, and expenses of these methods, new techniques have been developed. The selection of appropriate technology is dependent on several technical and non technical factors. The technical factors include assessment of the ability of the technology to reduce risk to human health and the environment, reliability and maintenance requirements for the technology, available infrastructure to support the candidate technology, ease of accessing the technology and associated services, risk to workers and public safety during the implementation of the technology, environmental impacts of the technology, the ability of the technology to meet regulatory acceptance, and obtaining of community acceptance [5,8]. The above mentioned technical factors should be integrated in a structured approach to assess the decision making process to select appropriate technologies. Table 3 lists a technology evaluation matrix developed by IAEA; this matrix provides a subjective ranking scale for each factor [6].

Recently, green and sustainable remediation (GSR) has emerged as a beneficial approach to optimize all phases of site

remediation, from site investigation to project closeout. Sustainable remediation was defined as a remedy or combination of remedies whose net benefit on human health and the environment is maximized through the judicious use of limited resources [9]. Environmental protection agency (EPA) identified five core elements of a GSR to assist with the selection of remediation technologies and approaches. These elements include [10];

1. Minimize total energy use and maximize use of renewable energy,
2. Minimize air pollutants and greenhouse gas emissions,
3. Minimize water use and impacts to water resources,
4. Reduce, reuse, and recycle material and waste, and
5. Protect land and ecosystems

In this work, the effort is directed to review advances in NORM and TENORM regulation in different countries, highlight recent trends in remediation technologies and identify knowledge gaps.

#### RECENT ADVANCES IN THE REGULATION OF NORM TENORM

IAEA had indicated that the regulators and operators are facing a variety of new challenges with NORM regulations, these challenges are attributed to the nature of traditional industries and their capabilities to deal with radioactive wastes [11]. Bad management of uranium mine and mills tail

ings can cause soil and groundwater pollution, to address this issue, regulatory bodies in different countries have issued national regulations. In 2011, UK have published guidelines from management of NORM in this guide, industrial activities that generate NORM were classified into two categories; activities that employ uranium or thorium and activities in which NORM presence is incidental. The concentration limit to include NORM as radioactive material was identified for different radionuclides, for example, if  $^{226}\text{Ra}$  exceed 0.5 Bq/g in solid or relevant liquid for first category, NORM will be treated as radioactive wastes [12].

In Canada, NORM management is not regulated by the Canadian nuclear safety commission, but it falls under jurisdiction of the provinces and territories. To eliminate cross jurisdictional boundaries among radiation protection standards, new guidelines were developed in 2000. In these guidelines, industries that generate NORM in significant amount to yield in radiation doses require the application of radiation protection practice were identified to include mineral extraction and processing; oil and gas production, metal recycling; forest products and thermal electric production, water treatment facilities; and tunneling and underground workings [13]. The guideline recommended the release of NORM from regulatory requirements if the associated dose was found less than 0.3mSv/y

In USA, NRC issued regulations to reduce emanations of radon from tailings, prevent the spreading of tailings through erosion, reduce contamination by seepage, and setting out requirements to perform risk assessment [14]. The regulation specified the utilization of passive disposal with a minimum life time of 200 years, the average maximum permissible radon concentration was recommended to be less than 5 pCi/g. The goals of the remediation activities were indicated within the regulation to achieve radon decay product concentration less than 0.03 working level and gamma radiation level not exceeding 20  $\mu\text{R}/\text{h}$  above background.

The Spanish limiting criteria for releasing NORM contaminated land are utilizing the following dose rate limit; doses should not exceed 300  $\mu\text{Sv}/\text{yr}$  (excluding radon doses),  $^{222}\text{Rn}$  concentrations in hypothetical future dwellings should not exceed 200 Bq/m<sup>3</sup>; and finally ALARA (as low as reasonable achievable) principles should be applied [15]. Michalik *et al.* concluded that irrespective of the final form of further European regulation, the evolution of the approach to protect the environment against ionizing radiation will force the radiological protection community to face the problem of how to quantify, in a convincing manner, the environmental impacts when an assessment is needed [16]. When there is no severe environmental impact it is enough for the provision of scientifically well justified evidence that the actual presence of radioactivity does not cause any effect on the environment.

**Table 1.** Activities that may lead to NORM contaminated residues and sites [2].

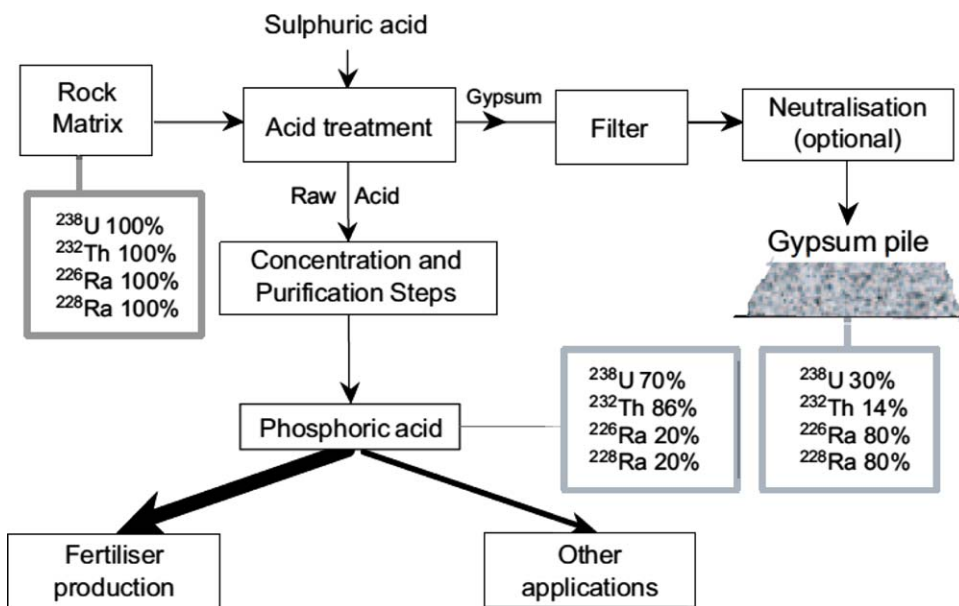
Mineral ores and extracted materials		Other processing/manufacturing
Copper	Titanium	Water treatment
Aluminum (bauxite)	Tungsten	Sewage treatment
Fluorspar	Vanadium	Spas
Gypsum	Zircon	Paper and pulp
Iron	Coal (and coal ash)	Ceramics manufacture
Molybdenum	Oil and gas	Paint and pigment manufacture
Phosphate	Geothermal energy	Metal foundry
Phosphorous	Thorium	Optics
Potassium		Incandescent gas mantles
Rare earth		Refractory and abrasive sands
Uranium		Electronics manufactures
Tin		Building materials

**Table 2.** IAEA radiological criteria to support remediation decision making [5, 6].

Band	Need for remediation actions	Acceptability of release	Range of annual doses
6	Remediation or prevent use	Not suitable for release,	> 100 mSv/a
5	Remediation or restrict use	Not suitable for release,	10-100 mSv/a
4	Remediation decisions based on justification/ optimization	May be released, but subject to regular review of situation	1-10 mSv/a
3	Remediation unlikely unless constrained	Released, but may need occasional review	0.1-1 mSv/a
2	Remediation unlikely to be necessary on the basis of radiological risks	Likely released, review is needed only if a problem becomes apparent	10-100 $\mu\text{Sv}/\text{a}$
1	No remediation necessary	Can be released without controls	< 10 $\mu\text{Sv}/\text{a}$

**Table 3.** IAEA remediation technology evaluation matrix [5, 6].

Evaluation factor	Exemplary	Acceptable	Unacceptable
Performance	Near 100% removal	Removes contaminants to desired limit	Mobilized or additional contaminant
Reliability	Near 100% reliable	Available without excessive down time	Unreliable
Maintenance	Minimal	Occasional	Unavailable suppliers or at great cost
Cost	Costs recoverable against credits	Cost within acceptable levels	Excessive cost
Infrastructure support technology	Not needed or fully available and already in place	Available	Unavailable or requires significant expense to provide
Availability	Well proven	Demonstrated and available in short time frame	Unproven/early in development
Risk	No risk to public or operators	Risk to public or operators within regulatory guidelines	More risk than if nothing done
Impact on environment	Clean and green	Little effect on overall ecosystem	Significant pollution/ damage
Regulatory acceptance	Exceeds regulatory standards	Meets regulatory standards	Fails regulatory standards
Community acceptance	Wholehearted acceptance without reservation	Acceptance with two way dialogue	Unacceptable



**Figure 1.** Radiocontaminant flow through phosphoric acid production [2]. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

Recently the Korean government had started to establish a regulatory framework for natural radiation including NORM/TENORM. The main motivation was the rapid economic growth in Korea that consume huge amount of raw materials, including NORM and TENORM [17].

#### RECENT ADVANCES IN RADIOLOGICAL ASSESSMENT OF TENORM

##### TENORM from Fertilizers Industry

To comply with radiation protection and environmental regulation there is a need to perform accurate and reliable assessment of the contamination and/or pollution extent. Fertilizers industry produces phosphogypsum, which is the principal TENORM waste product, the radio contaminant flow through the phosphoric acid production using wet sulphuric acid method is illustrated in Figure 1 [2].

Recently, the interest in evaluating the extent of NORM and TENORM in these industrial wastes and its radiological impact on human and the environment have been continued. This section is devoted to review these researches methodology and their most important findings. A new phosphogypsum reference material was produced and certified to assist in the validation of analytical methods and quality assurance of generated analytical results [18]. Sample preparation methodology, material homogeneity assessment, characterization campaign results and assignment of property values, and associated uncertainties were determined. The reference values and associated uncertainties for  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$ ,  $^{230}\text{Th}$ ,  $^{234}\text{U}$  and  $^{238}\text{U}$  were established based on consensus values calculated from analytical results reported by three National Metrology Institutes and five expert laboratories.

To evaluate the radiological impact of a phosphoric acid factory located in the south western Spain, the distribution and levels of radionuclides in the materials involved in the production process have been analyzed. The flow of radionuclides at each step was assessed and locations of possible radionuclide accumulation were identified. A set of samples collected along the whole production process were analyzed to determine their radionuclide content by both alpha particle and gamma spectrometry techniques. The radionuclide fractionation steps and enrichment sources have been located, allowing the establishment of their mass (activity) balances per year [19].

The radiological characterizations of disposed phosphogypsum were determined, the outdoor storage and transport radiological impacts and phosphogypsum usage as additive and filling materials in construction sectors were studied [20]. It was found that the mean activity concentrations of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  measured for the studied samples were in the range 436.0 125.0, 15.1 9.4 and 13.0 7.5 Bq/kg respectively. These values were concluded to fall below recommended safety limits for usability of phosphogypsum as aggregate in roads constructions sector in Turkey. Generic exposure scenario was used to evaluate the radiological impact on public members and workers, the results indicated that the evaluated mean annual effective doses were lower than the annual limit of 1mSv/y.

The chemical and radiochemical composition of a solid waste generated in phosphoric acid production plant were determined using analytical methods [21]. The analysis and leaching of samples containing  $^{210}\text{Pb}$ , and  $^{210}\text{Po}$  mixed with high activity concentration of uranium isotopes using gamma spectrometry and radiochemical methods showed a very low quantity of  $^{226}\text{Ra}$ . To evaluate the leachability of U isotopes,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ , sequential extraction method consisting of five operational defined fractions was used. The results indicated that the average leaching potential was 97.6% for  $^{238}\text{U}$ , 93.2% for  $^{210}\text{Pb}$  and 82.4% for  $^{210}\text{Po}$ . Moreover the study showed that  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  are leachable under extreme conditions, where U isotopes are more soluble.

During a preliminary survey at the area of an abandoned fertilizer plant, increased levels of radioactivity were measured at places, buildings, constructions and materials. The extent of the contamination was determined and the affected areas were characterized as controlled areas. After quantitative and qualitative determination of the contaminated materials, decontamination program was planned and performed step by step: the contaminated materials were categorized according to their physical characteristics (scrap metals, plastic pipes, scales and residues, building materials, etc) and according to their level of radioactivity. Depending on the material type, different decontamination and disposal options were proposed; the most appropriate technique was chosen based on technical factors, legal requirements, radiation protection standards, the opinion of stakeholders. After removing the biggest amount of the contaminated materials, an iterative process consisting of surveys and decontamination actions were performed in order to remove the residual traces of contamination from the area. During the final survey, no residual surface contamination was detected; some sparsely distributed low level contaminated materials deeply immersed into the soil were found and removed [22].

### TENORM from Water Treatment

In southeast Queensland, Australia, an assessment of TENORM generation during the treatment of water supplies was performed. Radioactivity concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$ ,  $^{222}\text{Rn}$ , and  $^{210}\text{Po}$  in surface water and groundwater samples were examined both pre and post treatment. The treatment of processes applied to surface water included

such as sedimentation, flocculation, coagulation, and filtration while cation exchange, reverse osmosis, and activated charcoals are used to treat groundwater. The secondary waste generated after treatment were identified to include sludges, exhausted ion exchangers, filtration media, back wash and wastewaters and their activity concentration were measured. The measured activity concentrations were used as input parameters to model the radiological impact of the secondary wastes. The results indicated that the water treatment practice in Australia does not pose a significant radiological risk [23].

### TENORM from Oil and Gas Industry

Oil and gas industries represent the main source for TENORM, the flow of radiocontaminants in different parts of the oil and gas production site and refining plants is illustrated in Figure 2 [2]. The distribution of radon gas and radiation exposure rates were evaluated in four natural gas treatment facilities in Syria [24]. The radiation exposure rates at the equipment area were found within the natural levels (0.09 0.1  $\mu\text{Sv/h}$ ) except for the reflex pumps where a dose rate value of 3  $\mu\text{Sv/h}$  was recorded. The concentrations of Radon in Syrian natural gas are in the range 15.4 1141Bq/m<sup>3</sup>; the concentration is higher in the associated natural gas than that of the non associated gas. The concentrating and pressurizing processes in the central processing facilities were found to be responsible for the detected high radon concentration. The lowest detected  $^{222}\text{Rn}$  concentration was found in the natural gas fraction equals 80 Bq/m<sup>3</sup>. On the other hand, maximum observed radon gas and its decay product concentrations were found to be high in the gas analysis laboratories (equal 458 Bq/m<sup>3</sup>). However, the reported levels in the studied stations were below the 1000 Bq/m<sup>3</sup> limit set by IAEA for radon chronic exposure [24].

Karen et al. highlighted the interest of both industry and regulators in identifying cost effective disposal alternatives that provide adequate protection of human health and the environment. One such alternative, currently allowed in Michigan with restrictions, is the disposal of TENORM wastes in nonhazardous waste landfills. The disposal of petroleum industry wastes containing  $^{226}\text{Ra}$  in nonhazardous landfills was modeled to evaluate the potential radiological doses and health risks to workers and the public. Multiple scenarios were considered in evaluating the potential risks associated with landfill operations and the future use of the property. The scenarios were defined to evaluate Michigan policy; sensitivity analyses were conducted to evaluate the impact of key parameters on potential risks. The results indicated that the disposal of petroleum TENORM wastes in nonhazardous landfills in accordance with Michigan policy and existing landfill regulations presents a negligible risk to most of the considered potential receptors [25].

The characterization of TENORM from the oil and gas industry in Egypt were overviewed, the average activity concentrations of  $^{226}\text{Ra}$  in scale and sludge samples were found to change between 5.9 and 68.9 kBq/kg (dry weight) in the waste samples from Gabal El Zeit field and Abu Rudeis fields, respectively. The mean activity concentrations of  $^{232}\text{Th}$  were 25.4, 2.6, and 7.2 kBq/kg and those of  $^{40}\text{K}$  were 1.3, 0.96, and 2.3 kBq/kg, for Abu Rudeis, granular Gabal El Zeit, and massive Gabal El Zeit [26].

### TENORM from Uranium Mining

As a benchmark for the old uranium mining sites remediation, the radiological health risk on the basis of dose assessment for naturally occurring uranium geochemical anomalies as a representative of Iberian Massif (Portuguese section) was conducted. Uranium, thorium and potassium radioactive series were measured in 52 samples taken from

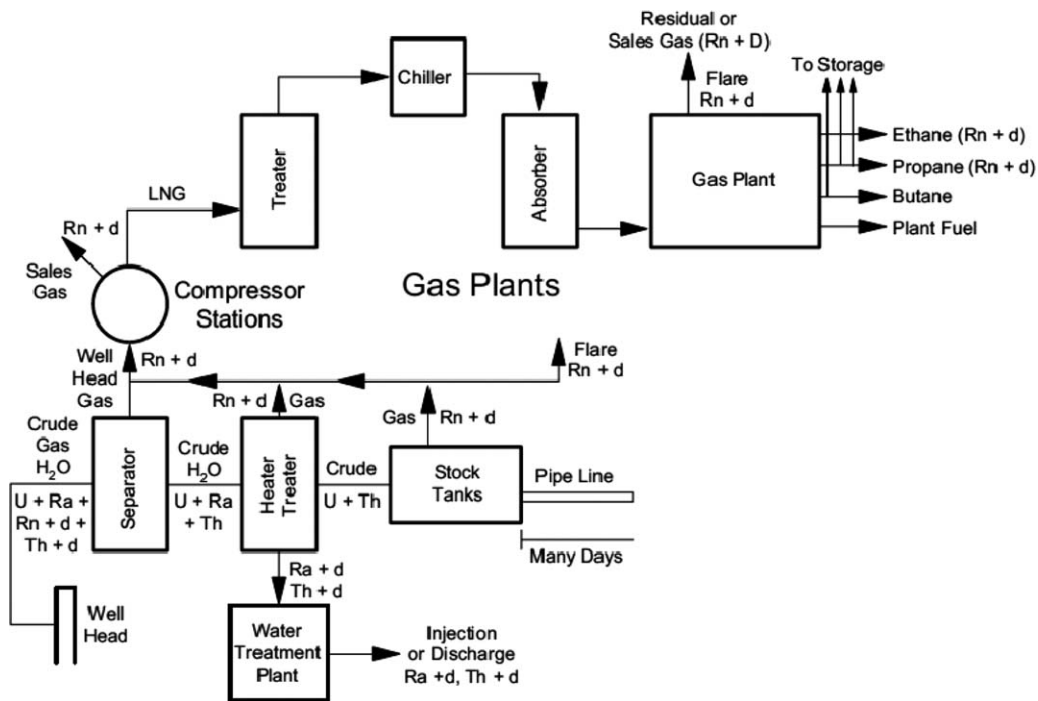


Figure 2. Radiocontaminants flow through gas and oil industry [2].

different environmental compartments: soils, stream sediments, water, foodstuff (vegetables) and air; external radiation was also measured through a square grid of 10×10m, with a total of 336 measurements. The results showed that some radioisotopes have high activities in all the environmental compartments as well as a large variability, namely for those of the uranium decay chain, which is a common situation in the regional geological setting. Isotopic disequilibrium is also common and led to an enrichment of several isotopes in the different pathways, as is the case of <sup>226</sup>Ra; maximum values of 1.76 Bq/L (water), 986 Bq/kg (soils), and 18.9 Bq/kg (in a turnip sample) were measured. On the basis of a realistic scenario combined with the experimental data, the effective dose from exposure to ionizing radiation for two groups of population (rural and urban) was calculated; the effective dose is variable between 8.0 and 9.5 mSv/y, which is 3-4 times higher than the world average [27].

Monitoring *in situ* leaching uranium mine groundwater, based on the monitoring of groundwater quality and the bottom value of groundwater pollution control was recently studied [28]. It was found that the operation of acid leaching mining area can lead to groundwater contamination with sulfate and nitrate. The determination of the current range of contaminant concentration is considered the basic data for the groundwater remediation.

#### REMEDICATION TECHNOLOGY

A wide range of remediation technologies is available to deal with contaminated sites, this section will highlight recent remediation researches applicable for remediating sites contaminated with NORM and TENORM such as passive barriers, pump and treat, electrokinetic, and bioremediation.

#### Barrier Technology

Permeable reactive barrier (PRB) is a class of passive engineering barriers that is installed to remediate contaminated groundwater. The basic idea of this technology is to place suitable material that has chemical reactivity toward

one or more contaminant in the contaminated groundwater path. As groundwater passes through the barrier, contaminants are removed by adsorption, exchange, oxidation reduction, or precipitation mechanisms of the barrier material [10]. Precipitation of phosphate minerals is one of the technological options used to remediate groundwater contaminated with uranium. The thermodynamic calculations for potassium and calcium uranyl phosphates, meta autunite and autunite showed that uranium concentrations will exceed the maximum contaminant level (0.13 μM for U) at any pH, unless phosphate concentrations is maintained higher than the sub μM levels typically found in groundwater [29]. Tokunaga *et al.* indicated that potassium uranyl vanadate can control uranium concentrations below regulatory limits in slightly acidic to neutral solutions. The calculations showed that maintaining uranium concentrations below the maximum contaminant level could be achieved by precipitating carnotite (K<sub>2</sub>(UO<sub>2</sub>)<sub>2</sub>V<sub>2</sub>O<sub>8</sub>) in oxidizing waters (pH 5.5-7) [29]. Experimental results for solutions at pH 6.0 and 7.8 were used to identify the optimum conditions to drop uranium concentration below MCL to provide contact within 1-5 days with oxidizing solutions containing 0.2-10 mM K, and 0.1-20 μM V(V).

Column experiments on remediation of acid seepage water from uranium containing debris using slaked lime and silica sand mixture as permeable reactive barrier have been conducted [30]. The results showed that the studied mixture gave good results; i.e., the utilization of one unit volume of the mixture can neutralize 108.3, 53.5, and 45.9 unit volume of acid seepage water (pH 3.12) with 1:5, 1:7, and 1:9 slaked lime to sand mass ratio. That work indicated that for contamination less than 0.05 mg/L uranium concentration in filtrate at pH < 6.5, the performance of uranium removal is good and fade removal performance for sulfate radical. The permeability of the mixture with 1:5 mass ratio was decreased from 10.9 m/d to 6.1 m/d, but the other two had no obviously change.

In Romania, there were three uranium mining areas, Banat region in southwest Romania, Apuseni Mountains in

the west, and Crucea in the north. The operation of these mines has led to pollution problems. The application of nanoparticles zero valent iron (nano Fe0) and nanoparticles magnetite (nano Fe<sub>3</sub>O<sub>4</sub>) for uranium removal from carbonate rich water taken from Lişava valley, Banat, Romania was performed. Nanoparticles were introduced to Lişava groundwater with a uranium VI. Liquid and nanoparticulate samples were collected and analyzed periodically over 84 days [31]. That study indicated that uranium 98% removal was achieved by nano Fe0 systems within 2 h of reaction, which reduce uranium concentration below EPA and WHO specified drinking water regulations. The results of X ray photoelectron spectroscopy analysis of the nanoparticulate solids confirmed the partial chemical reduction of uranium VI to Uranium IV, which is the most stable form. This result was attributed to the presence of Fe0 core that achieve near total removal of aqueous uranium even with the presence of competing ions. Over extended reaction periods (nearly 1 week) uranium VI was completely removed using nano Fe0.

Experimental studies using mine waters from Banat Romania area, in discontinuous single contact of phases was performed to select optimal reactive materials based on the distribution coefficient [32]. The results concluded the distribution coefficient of activated carbon type AC20G is the highest followed by that of apatite, while natural zeolites and zero valent iron had the lowest distribution coefficients. The fixed bed studies of these materials indicated that for an initial uranium concentration of 1.72 mg/L, the water could be decontaminated to less than 0.048 mg/L after 120 h.

Acid mine water from Straz pod Ralskem, Czech Republic, was remediated in by nanoparticles zero valent iron (nano Fe0). Toxicity of the remediated water was attributed to the presence of aluminum and sulphates in a high concentration, and the micro contaminants e.g. As, Cd, Cr, U, V, and Zn. Batch results showed significant decrease in pollutant concentrations due to pH enhancement and the decrease in oxidation reduction potential related to an application of nano Fe0. The contaminants were concluded to be removed by different mechanisms including cations precipitation in lower oxidation state, precipitation due to pH enhancement and coprecipitation with the formed iron oxyhydroxides [33].

Permeable reactive barriers for remediating acid mine drainage with low level uranium was studied [34]. In that work, 721 mine located in Jiangxi province were considered. The results showed that this remediation technique reduced uranium concentration to meet radioactive wastewater discharged standard, and acidity meets guide standard [34].

### **Pump and Treat Technology**

Pump and treat technology is one of the commonly used techniques for groundwater remediation. The development of unconfined groundwater flow and transport model using mesh free point collection method was presented by Mategaonkar *et al.* [35]. The developed models are coupled to get an effective simulation/optimization model for the groundwater remediation design using pump and treat technology. The simulation/optimization model was applied to the remediation design of an unconfined field aquifer polluted by Total Dissolved Solids (TDS) using pump and treat and flushing. The model provides an effective remediation design of pump and treat rate for the selected wells and costs of remediation [35].

Remediation of the Site Brook waterway was based on the need to remove contaminated floodplain soil and sediments while limiting the impacts on the surrounding ecological environment. The Site Brook remediation included soil and/or sediment excavation from wetlands, the wetland buffer zone, and upland areas. During the planning phase, characterization data were plotted and evaluated from historical surveys. In addition to the in stream surveys, workers

collected sediment samples using a hand auger from each grid where an elevated count rate greater than background was identified. For excavation of areas within the stream channel, a surface water diversion system was installed to convey stream flow around the excavation area and eliminate the flow of surface water through the excavation. A sump installed in the excavation area collected groundwater recharge into the excavation area and pumped it into a holding tank for radiological sampling and analysis prior to treatment via filtration and discharge to the Publicly Owned Treatment Works [36].

### **Electrokinetic Remediation**

Electrokinetic remediation is a relatively new physicochemical method used to mobilize ionic pollutants for removal in subsurface. Soil contaminated with organic and heavy metal pollutants could be remediated using this technology. The major drawback of this technology is the precipitation of reduced contaminants around the electrodes which implies the utilization of washing systems or adding augmenting fluids to prevent precipitates formation. The utilization of integrated bioelectrochemical system was proposed to overcome these drawbacks [37]. A microbial fuel cell was selected for studying the remediation of heavy metals such as uranium and chromium. The main factors that affect the performance of this technology were characterized. The results showed that the removal of hexavalent chromium is highly dependent on soil condition and the initial concentration.

To overcome the precipitation of reduced contaminants, improvements were carried out on the pilot scale electrokinetic equipment in Korean nuclear facility sites. These improvements include; immersion washing device, metal oxide separator, and circulation system. The aim from using the immersion washing device is cleaning the metal oxides from the cathode plate. The separator is used to remove the oxide particles below 0.075 mm. Finally, the circulation system is used to control pH and ensure waste solution movement. The pH in the cathode room is adjusted at 1.2, to prevent the generation of metal hydroxide and circulates waste solution to prevent the increase of metal oxides due to its stagnancy. After the remediation experiment for 25 days using improved pilot scale electrokinetic equipment, the removal efficiency of uranium from the soil was 96.8%, and its residual uranium concentration was 0.81 Bq/g. When the initial uranium concentration of the soil was 50 Bq/g, the electrokinetic remediation time required to remediate the uranium concentration below clearance concentration of 1.0 Bq/g was about 34 days. When the initial uranium concentration of soil was 75 Bq/g, the electrokinetic remediation time required to remediate below 1.0 Bq/g was about 42 days. When the initial uranium concentration of soil was 100 Bq/g, the electrokinetic remediation time required to remediate below 1.0 Bq/g was 49 days [38].

The treatment uranium leachate generated from the electrokinetic remediation was studied by using mixing and cohesion, precipitation, concentration, and filtration [39]. The leachate characteristics were performed and the results indicated that U concentration was 180 ppm. By using sodium hydroxide as a precipitant it was found that, the precipitation velocity was reduced and the precipitate particle size was increased. In several respects, sodium hydroxide was found to be more efficient as a precipitant than ammonium and calcium hydroxides. The results showed that a mixture of NaOH, alum, and magnetite (having weight ratio of 20:4:3) is optimal for filtration [39].

### **Biological Treatments**

Natural uranium and <sup>226</sup>Ra removal from contaminated waters by rhizofiltration was tested using *Helianthus annuus*

L. (sunflower) seedlings growing in a hydroponic medium [40]. The optimum age of the seedlings was studied and radionuclides removals using sunflower roots were tested. The results revealed that the seedlings induced the formation of precipitate that contains U and Ra. Contaminated water was remediated successfully using four week old seedlings for 2 days. 50% of the U and 70% of Ra were fixed in the roots, and the rest were detected in the precipitate [40].

Optimum conditions for penatron and soil mixing, and the soil pH of were studied using different bioremediations with soil contaminated by U and Ra. It was found that an optimum mixing ratio of penatron for bioremediation of uranium soil was 1%. Also, the optimum pH condition for bioremediation of soil contaminated with U and Ra was 7.5. Removal efficiencies from higher concentration of contaminated soil were reduced in comparison with those from lower concentration of soil. Meanwhile, the removal of uranium and radium in concrete by bioremediation is possible but the removal rate from concrete was slower than that from soil. The removal efficiencies of uranium and radium from soil under injection of 1% penatron at pH 7.5 for 120 days were 81.2 and 81.6%, respectively, and the removal efficiencies of uranium and radium from concrete under the same condition were 63.0 and 45.2%, respectively. Beyond 30 days, removal rates of uranium and radium from soil and concrete by bioremediation was very slow [41].

Uranyl ions biosorption on three types of alga: *Nostok linckia*, *Porphyridium cruentum*, and *Spirulina platensis* was studied. These ions were supplied either from a pure solution of uranyl nitrate, or after uranium ore leaching, or from the purification sludge. The retention degree versus contact time and afterwards the Langmuir and Freundlich biosorption isotherms of uranyl ions on the three alga types were investigated. The retention of contaminants ions on alga was proved through FTIR investigations. From the experimental data it was found that regardless of origin of uranyl ions, the retention degree on alga decreased in the series. *Spirulina platensis*  $\geq$  *Porphyridium cruentum*  $\geq$  *Nostok linckia* [42].

Geobacter mediated reductive immobilization of uranium is a novel bioremediation technique. While several reactive transport models have been developed to represent Geobacter mediated bioremediation of uranium, these models often lack the detailed quantitative description of the microbial process (e.g., biomass buildup in both ground water and sediments, electron transport system, etc.) and the interaction between biogeochemical and hydrological process. A recent study devoted to the development of a novel multi scale model by integrating a recent model on electron capacitance of Geobacter with a comprehensive simulator of coupled fluid flow, hydrologic transport, heat transfer, and biogeochemical reactions. This mechanistic reactive transport model accurately reproduces the experimental data for the bioremediation of uranium with acetate amendment. The proposed model captured significant contributing factors across time and space, thereby improving the structure and parameterization of the comprehensive reactive transport model. The global sensitivity analysis also provides a potentially useful tool to evaluate uranium bioremediation strategy. The simulations suggest that under difficult environments (e.g., highly contaminated with U(VI) at a high migration rate of solutes), the efficiency of uranium removal can be improved by adding Geobacter species to the contaminated site (bioaugmentation) in conjunction with the addition of electron donor (biostimulation). The simulations also highlight the interactive effect of initial cell concentration and flow rate on U(VI) reduction [43].

The utilization of phytoextraction for remediating soils contaminated with uranium has received increasing attention recently. The remediation of 10 cm soil depth having 1.5 kg dm<sup>-3</sup> density contaminated with Cs, Sr, and U by using

phytoextraction was studied. The annual removal percent for these contaminants were found 3.3, 5, and 9.3%, respectively which entailed the application of this method for 23, 10, and 7 years for each contaminant for the removal of 50% without the consideration of decay [44,45]. The feasibility of using citric acid, oxalic acid, nitrilotriacetic acid, and EDTA for phytoextraction of uranium tailings by Indian mustard was tested [46]. The tailings were mixed with garden soils at 1:3 ratio and four different chelators concentrations were added to the mixture. EDTA was found to produce maximum growth depression where nitrilotriacetic acid gives the minimum. The minimum growth inhibition observed was with nitrilotriacetic acid which was followed by oxalic acid, citric acid and finally EDTA. Another study devoted to investigate the effect of citrate, EDTA, and EDDS on the removal of radium from a granitic soil [47]. The removal process for all the studied chelators were found dependent on the substrate pH. The highest radium removal was obtained for citrate amendment at 50mmol/kg and the maximum removal was attained on the first day after amendment while using citrate will lead to delay till the fourth Day.

## CONCLUSIONS

This work aimed to highlight the scientific community interest in important topics that affect the selection of remediation technology. From this review the following conclusions could be drawn

1. Different materials have been tested for their potential use as reactive barriers to remediate uranium from contaminated groundwater, these include Potassium Uranyl Vanadate, slated lime and silica sand mixture, iron zero valent, magnetite, activated carbon, apatite, natural zeolites.
2. zero valent iron has received a great attention recently, but there is still a need to investigate how to improve the retention of inorganic contaminants in chemically complex environmental
3. Laboratory scale researches have been extensively performed but the pilot scale results were not adequately presented. So there is a need to study and/or model the effect of upscaling on the obtained experimental results.
4. Large uncertainties associated the fundamental process that governs bioreactor operation, i.e. transport and biocatalysis mechanisms, these uncertainties need to be addressed.

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