Towards the Application of Electrokinetic Remediation for Nuclear Site Decommissioning

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Abstract

Contamination encountered on nuclear sites includes radionuclides as well as a range of non-radioactive co-contaminants, often in low-permeability substrates such as concretes or clays. However, many commercial remediation techniques are ineffective in these substrates. By contrast, electrokinetic remediation (EKR), where an electric current is applied to remove contaminants from the treated media, retains high removal efficiencies in low permeability substrates. Here, we evaluate recent developments in EKR for the removal of radionuclides in contaminated substrates, including caesium, uranium and others, and the current benefits and limitations of this technology. Further, we assess the present state of EKR for nuclear site applications using real-world examples, and outline key areas for future application.

Keywords: Electrokinetic remediation; nuclear decommissioning; radioactive contamination

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1. Introduction

Anthropogenic radionuclide contamination arising from nuclear energy generation is a major land quality concern. Accordingly, many technologies exist for the remediation of contaminated materials and effluents at nuclear sites. These include bio- and phyto-remediation, thermal treatment, soil flushing, *in-situ* chemical oxidation (ISCO), ion exchange and excavation and encapsulation, among others.[1-5] Technologies that avoid physical removal or handling of the material (*in-situ*) offer significant benefits over *ex-situ* techniques, as *in-situ* approaches reduce the need to handle potentially contaminated materials meaning it is safer for workers. It also reduces the need to transport, store or dispose large quantities of materials, saving energy, waste disposal costs and reducing risk. These make *in-situ* technologies preferred on-site when safe and practical to do so. However, *in-situ* technologies must work effectively in a range of low- and high-permeability substrates (soils, sands, clays, *etc.*) as well as on or around infrastructure (pipework, plastics, concretes, steel, *etc.*). Many existing technologies struggle in low permeability substrates given limited water or reagent flow.

Electrokinetic remediation, EKR, is a technology with several potential advantages over established remediation methods. These include flexible set-up, low-energy requirements and, most importantly, an ability to work in low permeability substrates around (sub)surface infrastructure.[6] In contrast to soil flushing and similar techniques, EKR is effective in clays, silts and may be applied to varied tills, alluvial and lacustrine deposits, and loess, in addition to various process sludges and other materials. The wide ranging advantages of EKR techniques have driven a number of studies of EKR as a practical, on-site remediation technology including on active and legacy nuclear sites. Following an initial surge in papers in the late 1980s and early 1990s,[7] practical uptake of EKR as an "off-the-shelf" remediation technology has been limited however compared to other methods such as bioremediation, ISCO, *etc.* Research interest in EKR technologies has continued nonetheless (*e.g.* 123 papers were published in 2016, with "Electrokinetic remediation" either in the title or topic, as defined by Clarivate Analytics Web of Science platform). IIn addition, industry interest in EKR as an on-site and *in-situ* solution is re-emerging, and in the UK EKR is currently being examined as one of a range of technologies to address the UK's nuclear decommissioning legacy. This is under the UK-funded TRANSCEND project.[Note 1]

1.1 Electrokinetic Remediation, EKR

In EKR electrodes are implanted into a contaminated substrate and voltage applied. Under the effect of a low-energy direct current (DC) electric field, the movement of ions (electromigration), solid particulates (electrophoresis) and water (electro-osmosis) can be controlled and thus problem contaminants accumulated at desired points within a cell, usually around the electrodes. Water is also electrolysed within the cell with acid and alkaline fronts generated at the anode and cathode, respectively, shown in Figure 1, below. For the treatment of radionuclides this normally involves electromigration to the electrode of opposing polarity; *e.g.*, Cs^+ towards the cathode or $Tc^{VII}O_4^-$ towards the anode. Where radionuclides are sorbed to the surfaces of soil particulates, the combined effect from the electric field and pH gradient can help mobilise them into pore water, ensuring EKR remains effective for the remediation of a variety of radionuclide contaminants.[6]

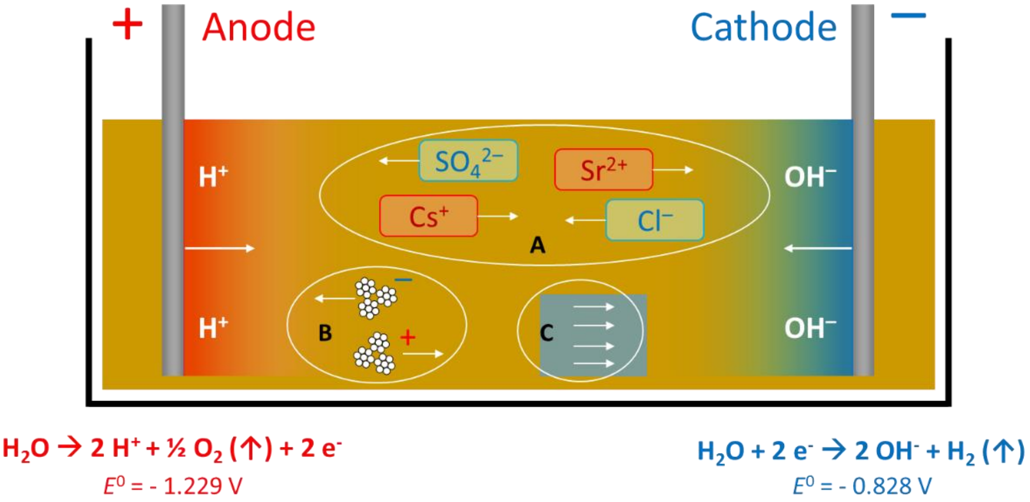


Figure 1 – EKR processes in a simplified cell, with pH fronts developing from the electrodes. **A**, electromigration of ions; **B**, electrophoresis of particulates (clays, *etc*.); **C**, electro-osmosis of (pore) water. Half-cell E^0 values are *vs*. the standard hydrogen electrode, SHE.

By driving contaminants out of solid wastes, EKR can be used to reduce the amount of solid material requiring disposal. Where liquid effluent generation is minimal, it may therefore be considered a *waste minimisation* technique. This aligns strongly with the preferred treatment routes outlined in the 'waste hierarchy' model of waste management, Figure 2, which outlines the preferred treatment options for hazardous wastes. This approach is widely employed in the UK nuclear sector and is a fundamental precept in radioactive inventory management.[8]

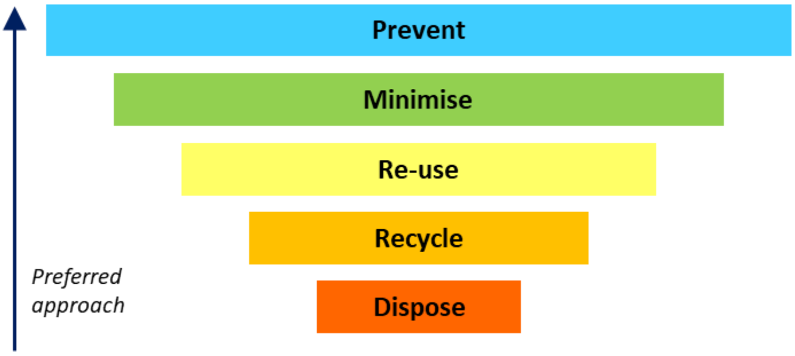


Figure 2 – The waste hierarchy. "Prevent", that is preventing waste from being generated where possible, is the preferred approach to managing radioactive inventories. Disposal is the least preferred option after waste minimisation, re-use and recycling have been considered.

The simplicity and low power use of EKR systems also ensure that they can easily be combined with other techniques. This adaptability has resulted in several studies in which EKR is combined with bio- or phyto-remediation (EKR-Bio or EKR-Phyto), nanoparticle delivery, dewatering, and others and there are a number of comprehensive reviews on the use of EKR for the remediation of organic[9-11] and other[12-14] pollutants. While EKR and electrochemical technologies for non-nuclear remediation schemes are a popular area of research,[15-21] there remains a lack of detailed and critical research and technology overviews directed towards using EKR in full-scale, sustainable applications on nuclear sites. We are unaware of any comprehensive review of EKR literature devoted specifically to nuclear legacy and remediation issues since 2009.[22] Recent years have also seen numerous advances in the application of sustainable remediation ("*a remedy or combination of remedies whose net benefit on human health and the environment is maximised through the judicious use of limited resources*")[23] approaches which despite the low energy, inexpensive and flexible nature of EKR equipment, has translated poorly into solutions at nuclear sites undergoing decommissioning. Given this knowledge gap we present our critical review on ongoing efforts and future directions for EKR for nuclear site remediation, applied sustainably.

1.2 Scope

While we aim to provide a comprehensive and accessible discussion of the current state of EKR technologies, we deliberately exclude detailed theoretical background on electrokinetic processes, which is sufficiently discussed elsewhere.[24] Our focus here is instead on three key questions, of more relevance to the application of EKR techniques than a re-examination of the first principles of EKR technology:

1. What developments have there been since 2009, the date of the last major review into EKR for nuclear sites,[22] for specific problem radionuclides?
2. How has EKR been applied and what are the primary challenges at selected nuclear sites with international importance?
3. How can EKR best be applied to address these challenges, especially within emerging sustainable remediation frameworks?

We survey recent, nuclide-specific advances in EKR (Section 2) and give a detailed examination of site-specific challenges at three international installations (Section 3). Finally, we discuss how EKR may be adapted to address future key challenges at these and other sites, particularly under sustainable remediation "drivers" (Section 4). We also include our estimate of the technology readiness level (TRL)[25] of EKR and other technologies, to reflect progress since 2009 towards large-scale implementation of this technology. Economic estimates are quoted in USD ($), calculated using a GBP (£) to USD ($) exchange rate of 1 to 1.3, and adjusted for inflation to 2019-2020 equivalent prices.

2. EKR Developments Since 2009 for Specific Problem Radionuclides

Contaminated materials on nuclear sites contain a complex mix of fission and/or activation products along with other non-radioactive contaminants, and deciding targets for remediation can be challenging. Korolev has previously[22] considered 'problem' radionuclides to include ^{90}Sr, ^{90}Y, ^{99}Tc, ^{137}Cs, ^{144}Ce, and actinides (U–Cm) for EKR at nuclear sites. Here, we refine this definition to include several 3*d* transition elements and lanthanides; Table 1. We also exclude radionuclides with short half-lives ($t_{1/2}$ < 1 year), as these do not contribute significantly to long-term radioactive contamination of affected sites on the timescales of site-wide remediation schemes.

Table 1 – Key radionuclides for EKR on nuclear sites. Half lives, decay modes, daughter products and fission yield data are compiled from Nuclear Data Services information, hosted by the IAEA. ec is electron capture, RN is radionuclide. Where multiple decay modes are known (*e.g.* ^{36}Cl; 98% β^- [^{36}Ar], 2% β^+ [^{36}S]) only the predominant mode (β^-) is shown.

Radionuclides	Half-life/y	Predominant decay mode [daughter created]	Comment
^{3}H	12.3	β^{-} [^{3}He]	Neutron activation of ^{6}Li (and minor fission product); widespread contaminant
^{14}C	5730	β^{-} [^{14}N]	Neutron activation of graphite or CO_2 coolant, present in organic or inorganic form
^{36}Cl	3.0×10^{5}	β^{-} [^{36}Ar]	Neutron activation of ^{35}Cl (mineral salts, *etc*.)[26]
^{41}Ca	$9.9\text{x}10^{4}$	ec [^{41}K]	Neutron activation of ^{40}Ca (cements, *etc*.)[27]
^{55}Fe	2.7	ec [^{55}Mn]	Neutron activation of steel construction materials[28]
^{60}Co	5.3	β^{-}, γ [^{60}Ni]	
^{59}Ni	$7.6\text{x}10^{4}$	ec [^{59}Co]	
^{63}Ni	100	β^{-} [^{63}Cu]	
^{90}Sr	28.9	β^{-} [^{90}Y]	Fission product (yield *ca*. 4.5%)
^{99}Tc	$2.1\text{x}10^{5}$	β^{-} [^{99}Y]	Fission product (yield *ca*. 6.1%)
^{125}Sb	2.8	β^{-} [^{125m}Te]	Fission products
^{126}Sn	2.3×10^{5}	β^{-} [^{126m}Sb]	
^{129}I	$1.6\text{x}10^{7}$	β^{-} [^{130}Xe]	
^{137}Cs	30.2	β^{-} [^{137m}Ba]	Fission product (yield *ca*. 6.3%)
^{152}**Eu**	13.5	ec [^{152}Sm]	
^{154}**Eu**	8.6	β^{-} [^{154}Gd]	
^{155}Eu	4.8	β^{-} [^{155}Gd]	
^{231}Pa	$3.3\text{x}10^{4}$	α [^{227}Ac]	Produced in fuel cycle
^{233}U	$1.6\text{x}10^{5}$	α [^{229}Th]	
^{234}U	$2.5\text{x}10^{5}$	α [^{230}Th]	
^{235}U	$7.0\text{x}10^{8}$	α [^{231}Th]	
^{236}U	$2.3\text{x}10^{7}$	α [^{232}Th]	
^{238}U	$4.5\text{x}10^{9}$	α [^{234}Th]	
^{236}Np	$1.6\text{x}10^{5}$	ec [^{236}U]	
^{237}Np	$2.1\text{x}10^{6}$	α [^{233}Pa]	

^{238}Pu	87.7	α [^{234}U]
^{239}Pu	$2.4\text{x}10^{4}$	α [^{235}U]
^{241}Pu	14.3	β^{-} [^{241}Am]
^{241}Am	432.2	α [^{237}Np]
^{244}Cm	18.1	α [^{240}Pu]

Significant developments in the EKR of uranium and, following the Fukushima-Daiichi nuclear power plant (FDNPP; Section 3.1) disaster and subsequent clean-up, radiocaesium have been reported since 2009. The surge in interest of radiocaesium remediation is particularly concentrated in Japan and South Korea, with a significant number of publications coming from these countries in recent years. We note also that ^{137}Cs is frequently examined in combination with ^{60}Co, an activation product of the neutron flux of stainless steel materials (*e.g.* pipework). It is a site-specific contaminant (*e.g.*, Hanford site, USA;[29] Winfrith, UK[30]) with a short half-life (5.3 years) that is produced in reactor containment materials. It is not normally present in ground contamination and tends to decay by the time general site remediation schemes start.

2.1 Radionuclide-specific remediation: caesium

Caesium-137 is a medium-lived fission isotope (^{235}U parent) with a half-life of 30.2 years and fission yield of *ca*. 6.3%, present in site wastes and from nuclear weapons testing, authorised nuclear facility discharges and major nuclear accidents. Construction waste-focused research in Korea has examined EKR applications for ^{137}Cs in artificially contaminated and crushed simulant TRIGA-reactor (TRIGA: training, research, isotopes, general atomics) concrete. For example, Kim *et al*. have reported one system in which Cs removal efficiencies of above 50% were achieved over 15 days with a 4 V.cm^{-1} gradient using > 0.01 M (acetic) acid as an electrolyte. Removal efficiencies in their system increased dramatically from 55% in 0.1 M acetic acid in a static cell to > 99% in 0.01 M acetic acid with a flow-system and pre-treatment with 3 M hydrochloric acid (Ti cathode, dimensionally stable anode (DSA)). This highlights the effect that intelligent process design, specifically *ex-situ* pre-treatment, can have on EKR removal efficiency.[31] Further work by Kim *et al.* showed pre-treatment of the same TRIGA-simulant concrete with 3 M sulfuric acid pre-treatment increased removal efficiency of ^{137}Cs from 52% (no pre-treatment) to > 99% (with sulfuric acid pre-treatment), using 0.01 M nitric acid as the electrolyte (15 days) and Ti cathode and DSA anode. Importantly, the radionuclide activity concentration detected in the remediated concrete was below the 100 Bq.kg^{-1} limit suggested 'safe' by the Korea Institute of Nuclear Safety. This enables these effluents to be disposed in municipal waste without further treatment, therefore aligning with the objectives of the waste hierarchy, Figure 2.[32] Effluent volumes were small (< 3 mL per gram of concrete remediated), and the effect of pre-treatment was to lower the pH of the concrete by partial dissolution/decomposition of the calcium carbonate.

However, removing one hazard (^{137}Cs) by introducing another (conc. acid) is clearly undesirable for on-site, *in-situ* remediation techniques, which are advantageous precisely because they are benign. A key development, therefore, towards dealing with contaminated concrete safely and cheaply on-site came recently from Parker *et al*., on showing that simply washing ^{137}Cs-contaminated concretes with aqueous KCl (0.4 M) increases the removal efficiency threefold from 19 (no KCl) to *ca.* 60% upon EKR (cathode: steel, anode: DSA).[33] The setup used by Parker *et al*. is shown in Figure 3.

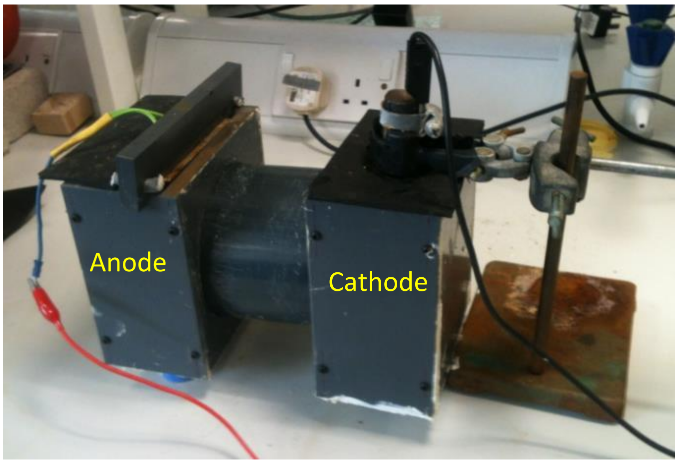


Figure 3 – EKR apparatus used by Parker *et al.* for remediation of ^{137}Cs-contaminated concrete. The anode is a platinum-titanium DSA, and cathode, right, is mild steel.

Enhancing EKR with a saline wash had been discussed previously[34] but until 2017 not applied on concretes. In this case the high strength KCl wash ion-exchanges the entrained Cs^+ ions for aqueous K^+ ions, mobilising them and enhancing the EKR effectiveness relative to EKR alone (*e.g.* without a KCl wash). An important caveat to this, however, is that high concentrations of competing ions retained in waste effluents could significantly hinder subsequent ion exchange-based clean-up processes for effluent treatment. Further work may therefore be needed to identify how high ionic strength solutions could be employed without inhibiting subsequent steps in the remediation process.

Radiocaesium in other wasteforms is also accessible by EKR. Soils surrounding the Korean Atomic Energy Research Institute, KAERI, have been intensively studied by Kim *et al.* since 2009.[35, 36] Sandstone is the primary rock type on which this and other Korean nuclear facilities are constructed, meaning EKR is effective in combination with pumping technologies. Indeed, a nitric acid electrolyte was most effective in the vertical EKR of $^{137}Cs^+$ contaminated (*ca.* 74 – 1643 Bq.kg^{-1}), acidic (pH 5.6) simulant material over between 20 and 50 days (nitric or acetic acid electrolyte, Ti cathode, DSA anode).[35] Removal efficiencies in the bespoke 10 L containment cell used ranged from *ca*. 57 to 94 % depending on soil pre-treatment, electric current (< 20 mA.cm^{-2}) and particle size, with approximately 2.5 mL of waste effluent generated per gram of remediated soil.[35] Scaling this apparatus fivefold to 50 L (Figure 4) resulted in no reduction in removal efficiency under nearly identical conditions (soil pH 6.4, 0.01 M nitric acid electrolyte, Ti cathode, DSA anode, 116 – 1186 Bq.kg^{-1}, 55 days), although more waste effluent (*ca*. 5 mL.g^{-1}) was generated.[36] Both techniques are amenable to electrodialysis with use of a selective ion-exchange membrane, resulting in ^{137}Cs removal efficiencies of up to 98% (21 days EKR, 600 mA, < 5.2 V).[37]

Figure 4 – The 50 L scaled set-up used by Kim *et al*. used to assess the scalability of the EKR cell to simulate remediation of contaminated KAERI sandstone. DSA is dimensionally-stable anode.[36]

In addition to work on Cs-contaminated soils and concretes, Kim *et al*. have examined EKR on incinerated waste ash (^{134}Cs- and ^{137}Cs-contaminated) and demonstrated a 75% removal efficiency of ^{134}Cs and ^{137}Cs after 7 days of EKR on the bench-scale with prior washing using nitric acid.[38] Removal efficiency in this 200 L washing/EKR apparatus was as high as 94% after 10 days EKR, using a 2 M nitric acid electrolyte.

2.2 Radionuclide-specific remediation: uranium

The five major isotopes of uranium (^{233}U, ^{234}U, ^{235}U, ^{236}U and ^{238}U) have long half-lives (≥ 10^5 years; Table 1) with *in-vivo* toxicity being the main hazard.[39] Enriched uranium wastes have the added hazard of being more radiotoxic. The remediation of uranium-contaminated soils remains an active area of research[40] and since 2009 EKR has achieved significant attention, including from Kim *et al*. and researchers at KAERI. Unlike ^{137}Cs, however, uranium is redox active and often co-precipitates (with CaO) as cathode plate-coating metal oxides, such as UO_2. These solids reduce the effectiveness of EKR and lower the removal efficiency, hindering pilot-scale *in*-situ remediation efforts.[41] To address this Kim *et al*. used a metal oxide particulate filter (75 μm) with pH control to successfully remediate a 50 L batch of contaminated (25 – 100 Bq.g^{-1} ^{238}U), slightly acidic (pH 6.2) soil (0.01 M nitric acid electrolyte, 28 V electric field over 28 – 49 days, Ti cathode, DSA anode). Under these conditions > 99% of uranium contamination in the soil was removed. Adding sulfuric acid to these solutions further inhibited metal oxide formation,[42] and aided recycling of contaminated electrolyte and uranium leachates.[43] Scaling the technology up from 50 to 800 L[44] (Figure 5) and even 1.2 tonne[45] mixing silos had little to no negative impact on the EKR removal efficiency of ^{238}U. Removal efficiencies of *ca*. 94% (30 days EKR, 20 V system, water or nitric acid electrolyte, Ti cathodes, DSA anodes) and up to 83% (40 days EKR) were achieved for the 800 L and 1.2 tonne systems, respectively.

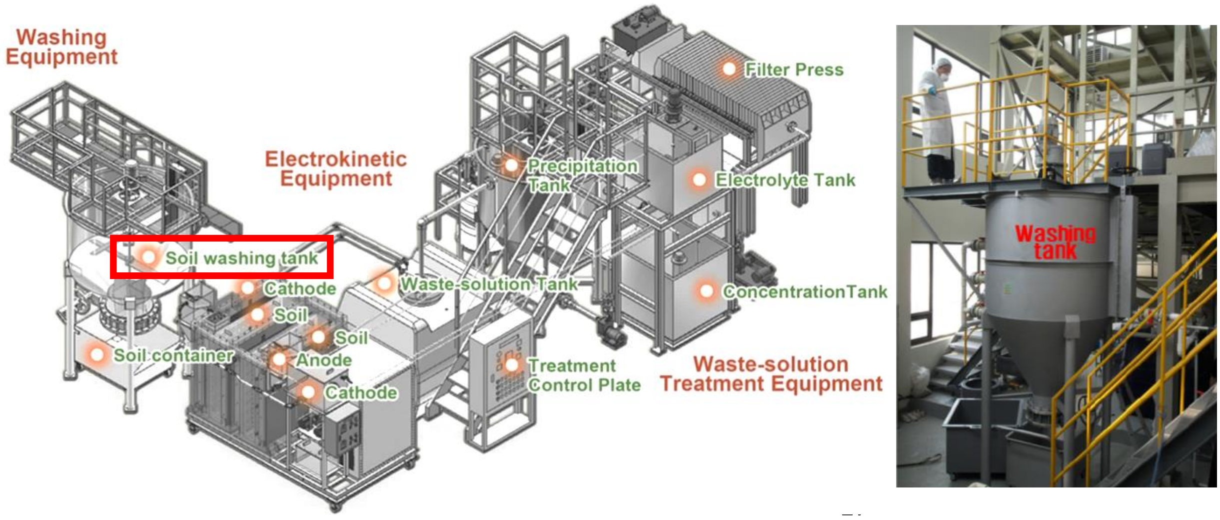


Figure 5 – Schematic of the set-up used by Kim *et al*. on the 800 L scale EKR process on contaminated KAERI material, with washing drum highlighted on the right.[44] The drum is several metres tall so is a significant improvement from laboratory scale.

An emerging area of EKR research is uranium removal from concretes and other building aggregates and developing flowcharts for material processing. Kim *et al*. have shown that remediating the floor of a former uranium processing facility (containing epoxy and urethane binders, cements, aggregate, *etc*.) can be enhanced by ball milling the material prior to treatment.[46] Although EKR was not performed on these materials the authors suggested a process flowchart, hypothesising a 70% reduction in waste volumes compared to other techniques[47] where EKR was applied in tandem with *ex-situ* milling. Yurchenko *et al*. have taken this one step further, by applying EKR to reinforced concrete (Figure 6) from a uranium processing facility in Moscow, contaminated with ^{235}U and ^{238}U.[48]

Figure 6 – Examples of the concrete examined by Yurchenko *et al.*,[48] complete with reinforced steel pins used in construction. X-ray (Rietveld) analysis indicated α-quartz, orthoclase and plagioclase feldspar, calcite and boltwoodite – $H(K,Na)(UO_2)(SiO_4)\cdot 1.5(H_2O)$ – phases in the concretes.

Here, crushing of 98 kg of authentic concrete building materials (complete with reinforcing steel pins and other concrete building supports), washing (sodium carbonate electrolyte) and applying the EKR process (0.22 $A.cm^{-2}$) over 30 days resulted in activity dropping by 8.2 MBq in the remediated concrete, a reduction of 95%. Only small amounts of waste (3.5 – 3.8 mL effluent per g of remediated concrete) were generated, an important step towards potential scale-up and aligning with the waste hierarchy (Figure 2). Gravels (> 10 mm) may also be treated by EKR. As 20 – 30% of soil surrounding the Korean KAERI facility is gravel,[49] Kim *et al*. were motivated to remediate the ^{238}U levels from *ca*. 10 $Bq.g^{-1}$ to below 0.43 $Bq.g^{-1}$, the disposal limit for landfill.[50] Depending on particulate size, the authors proposed a processing flowchart including ball mill crushing or soil washing, followed by electrodialytic (pump) EKR on the resulting aggregates. Removal efficiencies after EKR for 20 days (20 V, 200 L reactor, nitric acid electrolyte) were as high as 83%, with an average of 37% depending on pre-treatment processing.

2.3 Radionuclide-specific remediation: other nuclides (^{99}Tc, Eu, Pu, *etc.*)

Other, easier-to-detect radionuclides have also been targeted through EKR in recent years. For example, Agnew *et al*. demonstrated for the first time in 2011 the on-site EKR of Pu-contaminated soils at the Atomic Weapons Establishment (AWE) Aldermaston site in the UK.[51] This pilot study used a 10-electrode array (Figure 7; 19.2 V, 30% citric acid electrolyte, steel electrodes) implanted into *ca*. 4 tonnes of soil and monitored the gross alpha activity (*e.g.* Pu and Am) after EKR for 60 days. The study showed *ca.* 60% of monitored soil could be reclassified to the level of 'exempt' waste, based on the exemption limit of < 0.4 $Bq.g^{-1}$ above background alpha and beta contamination relevant at the time of the study.[51, 52]

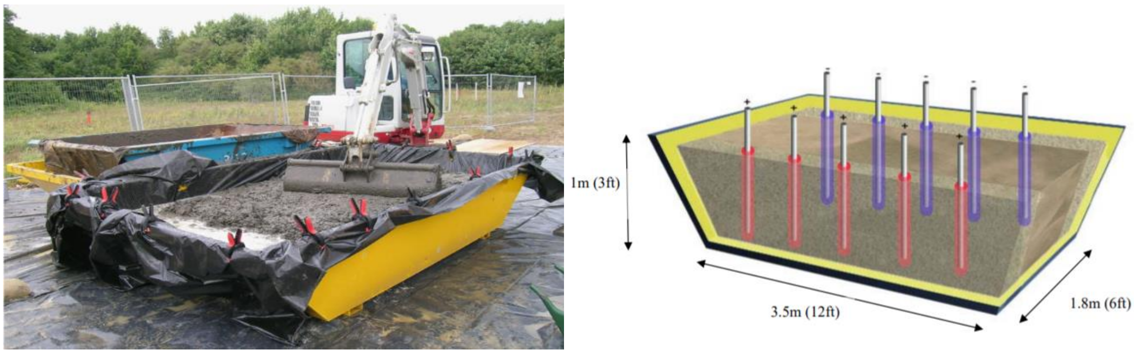


Figure 7 – Left, *ex-situ* processing of plutonium contaminated waste by Agnew *et al*. at the Atomic Weapons Establishment (AWE), UK, plus scale and electrode arrangement used on the right.[51]

The estimated cost (2011) of remediating this waste was *ca*. \$2700 per m^3 (2019 USD; £2054), less than *ca*. \$7830 per m^3 (2019 USD; £6054) for conventional excavation and disposal ('dig and dump').

Irradiated nuclear material may also be remediated electrochemically (but not, specifically, electrokinetically), as demonstrated by Bespala *et al*. in the treatment of spent graphite from the Zheleznogorsk nuclear reactor in Russia.[53] By using the contaminated graphite rods as electrodes, Bespala *et al*. demonstrated that applying an electric field of varying voltages (1 – 32 V) in nitric or sulfuric acid or DI water electrolytes removes ^{60}Co, ^{137}Cs, and $^{152/154}$Eu from the electrodes. Potassium permanganate added to selected electrolyte baths also allowed complete dissolution of the graphite electrodes upon application of electric current.

Finally, Valdovonis *et al*. have shown that EKR of ^{24}Na and ^{99m}Tc (as analogues for the longer lived isotopes ^{22}Na and ^{99}Tc, a difficult-to-measure nuclide) is effective in either removing ($^{24}Na^+$) or migrating and reducing (^{99m}Tc, as $^{99m}Tc^{VII}O_4^-$) present in Mexican phaeozemic (*cf*. mollisolic)[54] soil in the presence of scintillation fluids. These fluids were adsorbed onto the soils during the 1970s and 80s as an interim storage method.[55] Removal efficiencies after 4 hours were highest near the cathode (Ti rod) for $^{24}Na^+$ (*ca*. 72% from initial), and the anode (DSA) for ^{99m}Tc ($^{99m}TcO_4^-$; 61% removal from initial after 4 hours EKR) with a *ca*. 32 V system.

2.4 Summary of EKR targets, efficiencies and parameters in post-2009 experiments

Summary data from these and the other experiments outlined here is given in Table 2, below.

Table 2 – Summary of EKR experiments described in Section 2. Multiple experiments are often reported in the same reference, and selected data refer to experiments with the highest quoted removal efficiency for a given element or radionuclide. Abbreviations: n.r. is not reported, conc. is concentration, aq. is aqueous, soln. is solution, and DSA is dimensionally-stable anode (two or more metal oxides (*e.g.,* $Pt^{IV}O_2$ and $Ti^{IV}O_2$) grafted onto a corrosion-resistant mesh). Ultima Gold XR® is a branded reagent mixture commonly used in liquid scintillation counting analysis.

Elements or *RNs*	Wasteform	Voltage	Current	Electrode material	Time/d	Highest quoted removal efficiency	Bench, intermediate, or pilot scale?	Electrolyte	Ref.
Cs, Co	crushed concrete	4 V.cm^{-1}	500 mA	cathode: Ti anode: DSA	15	99+% for both	bench	Acetic acid (1 L, 0.01 M)	31
^{60}Co, ^{137}Cs	crushed concrete	n.r	20 mA.cm^{-2}	cathode: Ti anode: DSA	15	99.6% ^{137}Cs 99.7% ^{60}Co	bench	Nitric acid (0.01 M)	32
^{137}Cs	concrete	60 V	35 mA	cathode: steel anode: DSA	37.5	60%	bench	Aq. KCl (0.4 M)	33
^{60}Co, ^{137}Cs	washed soil	n.r.	20 mA.cm^{-2}	cathode: Ti anode: DSA	50	99.9% ^{60}Co 94.3% ^{137}Cs	bench	Nitric and acetic acid	35
^{60}Co, ^{137}Cs	washed soil	n.r.	15 mA.cm^{-2}	cathode: Ti anode: DSA	55	95.8% (both)	intermediate (50 L)	Nitric acid (0.01 M)	36
^{137}Cs	washed soil	< 5.2 V	600 mA	n.r.	21	98.2%	bench	*With ion exchange membrane*	37
^{134}Cs, ^{137}Cs	ash	10.5 V	2.2 A	n.r	7.2	74.7% ^{134}Cs 75.5% ^{137}Cs	bench	Nitric acid (2 M)	38
		n.r	n.r.		10	94.0% ^{134}Cs 93.9% ^{137}Cs	intermediate (200 L)	Nitric acid (conc. n.r.)	
U	soil	28	15 mA.cm^{-2}	cathode: Ti anode: DSA	49	99.0%	intermediate (50 L)	Nitric acid (0.01 M)	41
^{238}U	washed soil	20	13 mA.cm^{-2}	cathode: Ti anode: DSA	30	93.9%	pilot (800 L)	Nitric acid (3 M)	44
U	washed soil	n.r.	18 mA.cm^{-2}	cathode: Ti anode: DSA	40	83.3	pilot (1.2 tonnes)	Nitric acid (conc. n.r.)	45
U (various)	concrete	n.r.	0.22 A.cm^{-2}	n.r.	30	n.r.	bench	Aq. Na_2CO_3	48
U	crushed gravel	20 V	200 A	n.r.	20	83%	intermediate (400 L)	Nitric acid (conc. n.r.)	50
Pu, Am	soil	19.2	n.r.	steel	60	n.r.	pilot (4 tonnes)	Aq. citric acid (30% soln.)	51
^{152}Eu, ^{154}Eu (+ ^{60}Co, ^{137}Cs)	reactor graphite	31.8 (max.)	< 0.3 A.cm^{-2}	reactor graphite	n.r.	n.r.	bench	Mineral acids (< 8 M)	53
^{24}Na, ^{99m}Tc	soil	n.r.	1 mA	cathode: Ti rod anode: DSA	0.17	71.8% ^{24}Na 61.0% ^{99m}Tc	bench	1:1 Ultima Gold XR:water	55

The removal efficiency of selected elements from a variety of wasteforms is high (often 90%+). Although many studies are limited to bench-scale, our review highlights that progress is being made towards scaling EKR for use on-site. Pilot-scale studies up to tonnes of material show little to no loss of EKR effectiveness and although the current state-of-the-art is *ex-situ*, demonstrating successful pilot-scale viability is vital to ensuring EKR can be used effectively on site.

3. EKR at Nuclear Sites of International Importance

As noted, the wide scale deployment of EKR at complex nuclear sites, such as FDNPP (Japan), the Hanford site (US), and Sellafield (UK) is currently lacking. Contamination at these sites often exists for decades in many wasteforms, meaning remediating these sites is expensive, technologically difficult and hazardous. It is typically only governments that possess the resources and logistical ability to successfully remediate such sites, managed through governmental bodies and subsidiaries. These include the Nuclear Decommissioning Authority (NDA) in the UK, the US Department of Energy (DoE) and the Tokyo Electric Power Company (TEPCO) in Japan, and subsidiaries. As governments are ultimately responsible to taxpayers, remediation efforts are always high on the political agenda, providing substantial political and economic pressures to ensure remediation is both successful and cost effective. Here we outline three of the most challenging international nuclear legacy sites, and our assessment of the opportunities and challenges that EKR schemes could face if applied.

3.1 Fukushima-Daiichi Nuclear Power Plant (FDNPP) and exclusion zone, Japan

Following meltdown at the FDNPP, significant quantities (*ca*. 3.3 PBq) of ^{137}Cs and ^{90}Sr were released into the atmosphere. Much of this remains trapped within the 30 km exclusion zone, and the total bill estimated in 2016 of between $50-150 billion.[56]

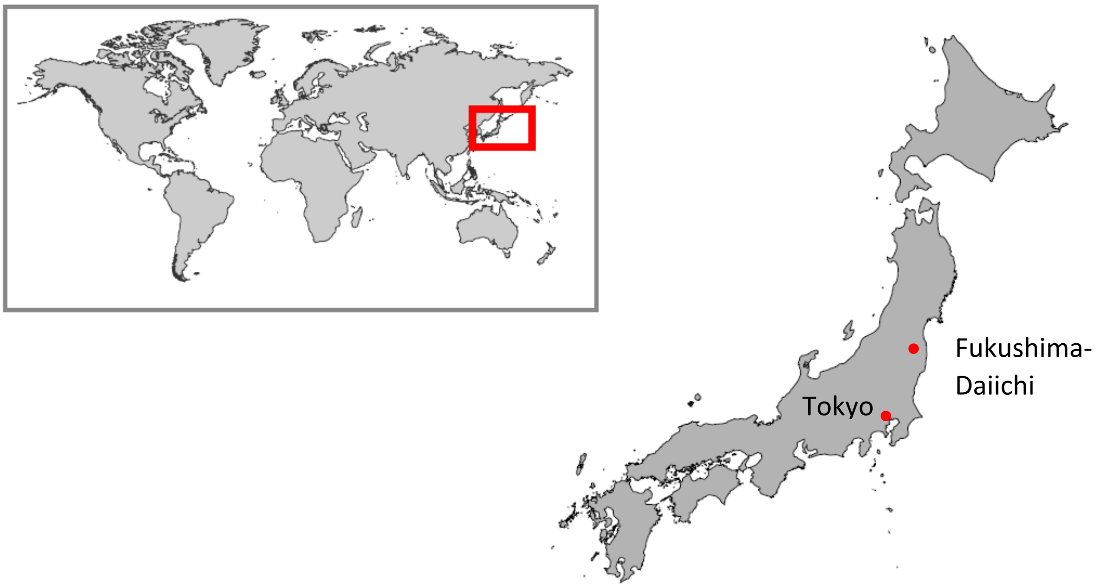


Figure 8 – Global and national location of the FDNPP in Eastern Japan. The figure omits the Okinawa Islands to the south of Kyushu.

The current remediation method, topsoil removal ('dig and dump') generates considerable soil (estimated 20 million m^3 from 13,000 km^2 of land, 2019)[57] waste which will need to be remediated if TEPCO and the Government of Japan are to fully implement their final disposal plan. Currently this involves constructing a permanent storage bunker within the Fukushima prefecture by 2045 but projections (2018) indicate this target will be missed without significantly enhanced volume reduction measures. Local opposition in all previously consulted sites has been also considerable and widespread, further complicating remediation efforts.[58]

There two noteworthy areas of the Fukushima exclusion zone, an organic-rich, clayey-silt with significant vegetation,[59] and an alluvial Quaternary-era sand terrace. The clayey-silt contains significant amounts of phyllosilicate micas (such as high cation-exchange capacity illite and biotite) that retain entrained $^{137}Cs^{+}$ within clay matrices.[60] Much of this contamination is concentrated in the top 5 cm of topsoil and is unaffected by rainwater flow,[61] meaning soil flushing, ion exchange, and similar technologies are ineffective at remediating these; other remediation techniques must be applied. The second area, the Quaternary era sandy terrace, underlies the FDNPP itself and contains significant groundwater contamination resulting from the core flushing immediately after containment failure. More than 1 million tonnes of this caesium and tritium contaminated waste water is currently stored at Fukushima (from the Advanced Liquid reProcessing System (ALPS) and Simplified Active water Retrieve and RecoverY (SARRY) systems) in corrodible water storage tanks.[62] TEPCO estimates that at current rates tank space will be exhausted by the summer of 2022 and has contingencies to discharge contaminated liquid wastes into the Pacific Ocean.

To date, we are not aware of any successful, site-scale attempts to remediate either the soil or groundwater within the Fukushima exclusion zone. Although an "ice-wall" of frozen brine-saturated soil (estimated cost of \$350 – 400 million) was deployed to limit the flow and volume of contaminated groundwater, the success of the scheme is debatable and many international news agencies have carried articles questioning its effectiveness.[NOTE 2] EKR technology (EK fencing,[63] EKF, where flow of contaminants is inhibited by application of electrodes, Figure 9) has been proposed to limit the spread of groundwater[64] or remediate contaminated seawater,[65] but we have not been able to find any primary literature to suggest these proposals were successful. Although small scale, limited evidence also exists suggesting that EKR of mountainous slopes contaminated with ^{137}Cs may be possible.[66]

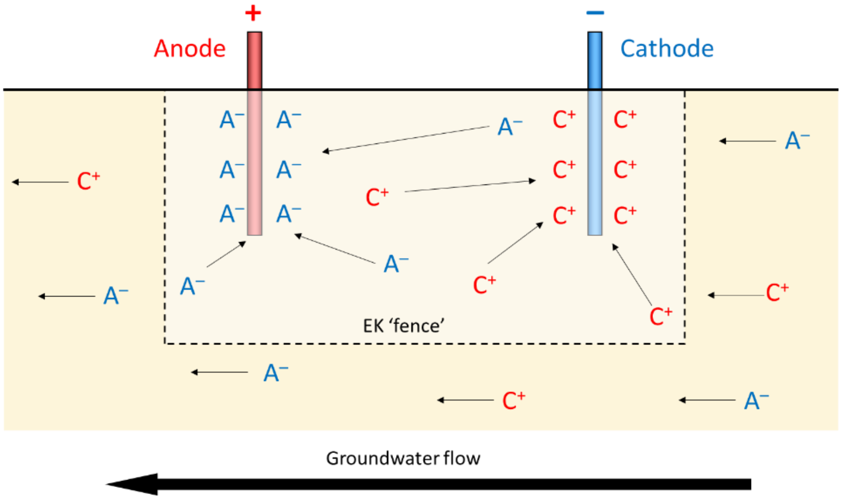


Figure 9 – Simplified schematic illustrating EK fencing, EKF. A^{-} and C^{+} are generic anions or cations, respectively.

Any successful remediation technology must address several key challenges at Fukushima. It must be versatile, and work in both low-permeability clays (*e.g.* forest soils), and higher permeability sandstone deposits (that underlie the FDNPP).[67] Techniques that work only in high-permeability substrates are not applicable for site-wide use. Secondly it must be cheap, and while EKR is often considered a low cost technique (Section 1.1), one of the key limitations of the EKF proposals by Lambda Consult are projected costs of \$47 – 64 million (£36 – 49 m) in capital outlays and \$2.8 – 5.6 million (£2.1 – 4.3 m) in annual running costs. Although a smaller pilot project (6 months, \$278k/£213k) was proposed we have been unable to confirm if this was funded. Thirdly, although much $^{137}Cs^{+}$ remains entrained within clay lattices,[68] clayey soils rich in organic carbon, such as those within the Fukushima exclusion zone, often contain significant micro-aggregated regions in which ^{137}Cs preferentially resides. Koarashi *et al.* have previously demonstrated that between 69 – 83% of $^{137}Cs^{+}$

in Fukushima soils resides in micro- or sand-sized-aggregate particulates, which have much higher extractability than clay regions.[69]

Given the on-going and controversial plans currently being employed by TEPCO for FDNPP remediation, we suggest that EKR may be beneficial in targeted, small-scale testing in authentic contaminated Fukushima soils. We have begun to make progress towards this goal, with work in our laboratories showing significant Cs^+ mobilisation from a Fukushima simulant soil upon application of a low-energy EK field.[70] Here, we demonstrated removal efficiencies of 80+% for Cs, over timescales of up to 45 days in cells with low power requirements ($<$ 1 kWh and $<$ 1 $V.cm^{-1}$). We are also aware of work at the University of Leeds, UK and the Korean Advanced Institute of Science and Technology (KAIST), South Korea, to develop new materials to selectively extract entrained Cs^+ in clay interlayers such as those present at Fukushima.[71] These studies should highlight the likely efficacy of EKR to the FDNPP in more detail, and we eagerly anticipate such results.

3.2 Hanford Site, Washington, USA

The Hanford Site (Figure 10) is the largest and oldest nuclear site in the US, having provided much of the plutonium enrichment capability for the USA during the 1940s and 1950s.

Figure 10 – Global and national location of the Hanford site, Washington state, USA.

The site has widespread contamination (^{60}Co, ^{90}Sr, ^{99}Tc, ^{129}I, ^{3}H, plus ^{106}Ru, ^{144}Ce, ^{147}Pm, various U and Pu isotopes and organics including CCl_4)[72, 73], although wastewater tank leakage remain most pressing. This includes significant ^{137}Cs contamination in some areas greater than 4 $KBq.g^{-1}$, Figure 11.[74-76] Remediation is extremely expensive, and recent estimates (2019) target a nuclear end-state of 2130 at a cost of between \$323 and \$677 billion (*ca*. £250 to £525 billion).[77]

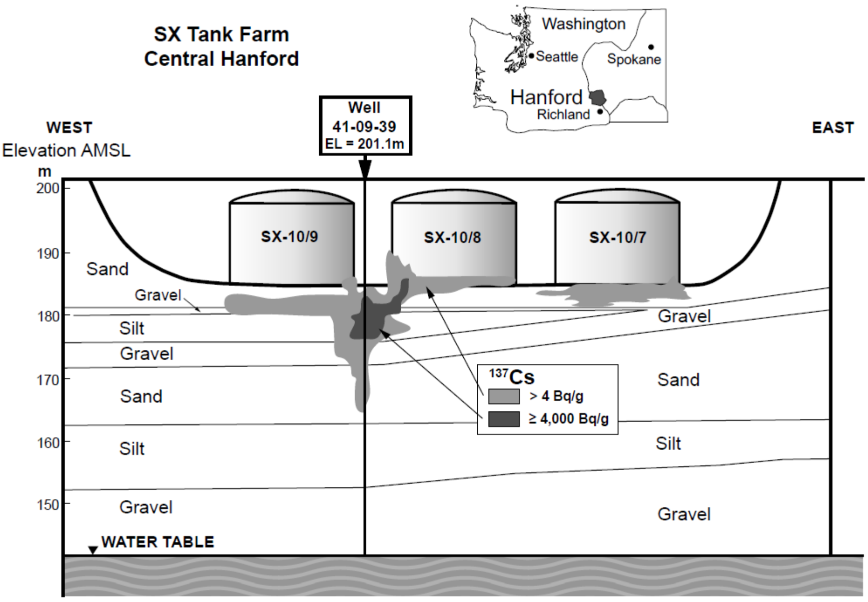


Figure 11 – Extent and levels of subsurface ^{137}Cs-contamination below three exemplar tanks at the Hanford site. The figure is redrawn from McKinley *et al*.[75]

The surficial geology of the Hanford site is primarily unconsolidated, high-permeability sands and gravels.[78] This means that unlike the clayey soils surrounding the FDNPP, a range of remediation techniques have been tested or are currently applied on the Hanford site at varying scales.[79] These include the injection of an apatite-rich barrier to limit ^{90}Sr influx into the Columbia River,[80] ongoing pump-and-treat systems,[81, 82] simple excavation,[83] and the bioremediation of residual organic contaminants in groundwater.[84, 85] EKR on the Hanford site has received less attention, although a small number of studies have examined EKR on Hanford-relevant materials to date.

In 1994, Buehler *et al*. artificially spiked authentic Hanford sediment with either a 1.85 MBq ^{137}Cs, or 0.74 MBq ^{137}Cs/1.85 MBq ^{60}Co solution, and applied an EKR treatment whilst monitoring γ-activities over 200 hours. Under a 200 V electric field (22.8 cm cell; 7 V.cm^{-1})[29] both ^{137}Cs and ^{60}Co migrated a small but significant distance (10 cm) towards the cathode, leaving much of the soil radiometrically free of contamination. In 2015, Jung *et al*. illustrated (Figure 12) that EKR on ^{133}Cs-contaminated Hanford site soil for 68 days can reduce the Cs concentration by up to 55%. Cs removal by EKR was more effective in silty-clayey fractions (average 51%) than sandy ones (average 38%), primarily from the cathodic region of the tested cell.[86]

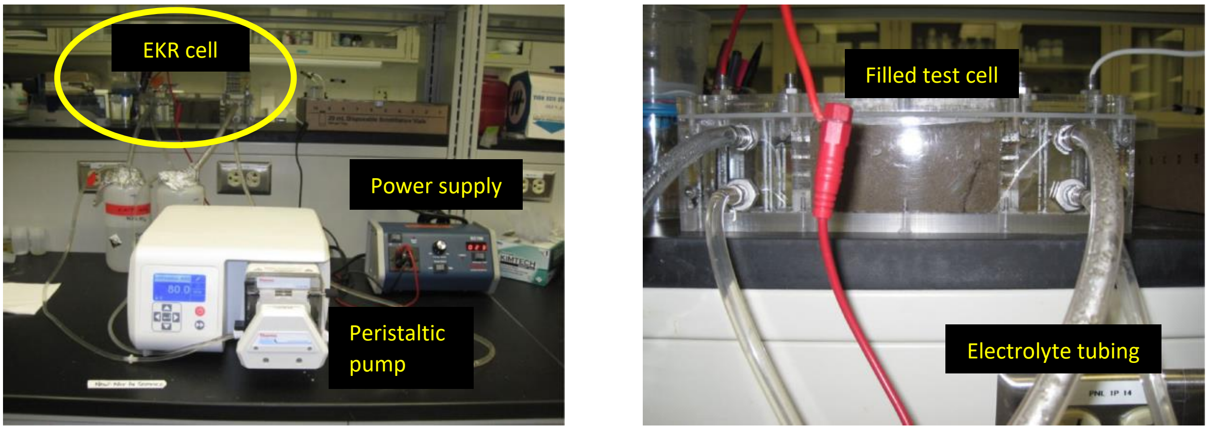


Figure 12 – Left, the set-up used by Jung *et al*, and right, magnified view of the clear Perspex® container (width = 25 cm) used for the EKR experiments.[86]

A key limitation with both of these studies is the size of the experiments; all cells were less than 25 cm in length. Owing to its comparative technological immaturity to other techniques, we suggest that the niche of EKR at Hanford is to supplement existing technologies. One example is the growth of the aforementioned apatite permeable reactive barriers (PRBs),[80] where apatite (calcium phosphate) precursors were injected into target sites along the Columbia Riverbank and ^{90}Sr measurements taken over time. EKR has been previously used to force-migrate and focus reactants into specific points along a remedial cell and we believe a similar approach could be of interest here, where existing injection methods prove ineffective.[87] Calcium[88] (and by extension strontium) and phosphate[89] ions are known to be mobile in soils under the effect of an applied electric field, and with careful electrode placement the EKR-enhanced growth of *in-situ* generated PRBs remains into difficult-to-inject materials could be an interesting prospect to explore.

3.3 Sellafield, Cumbria, UK

The third of the selected international sites discussed here is the Sellafield industrial complex in NW England, UK, the largest nuclear complex in Europe.[90] As much as 12 million m^3 of soil may be radioactively contaminated, including 1,600 m^3 of highly hazardous intermediate-level waste.[90] Significant contamination also resides in the site groundwater, which shows maximal activity concentrations of ^{137}Cs, ^{90}Sr and ^{99}Tc of 129, 84,000, and 71 Bq.L^{-1}, respectively.[91] These values are above or very close to World Health Organisation (WHO) guidelines for safe drinking water (10 Bq.L^{-1} for ^{90}Sr and ^{137}Cs, and 100 Bq.L^{-1} for ^{99}Tc).[92] Tritium is also a significant contaminant at the Sellafield site, with concentrations in analysed boreholes consistently above WHO guidelines (10,000 Bq.L^{-1}). In some cases concentrations can exceed the guideline limit by a factor of over 25 (255,000 Bq.L^{-1}).[92] We discuss these difficult-to-measure (DTM) radionuclides further in Section 5.

Targeting a nuclear end-state of 2120, the UK Government estimates decommissioning costs of *ca*. $117 billion (£91 billion; 2018),[93]although this has risen substantially in recent years (2009 costs were $80 billion/£62 billion, and in 2012, $102 billion/£79 billion)[94] and may in fact be as high as $210/£163 billion (2015, est.).[95] Decommissioning activities at Sellafield dominate the UK nuclear legacy estate, accounting for over 75% of remediation expenditure, Figure 13, in 2018-2019.[96] This has risen from 74% in 2015-2016.[95]

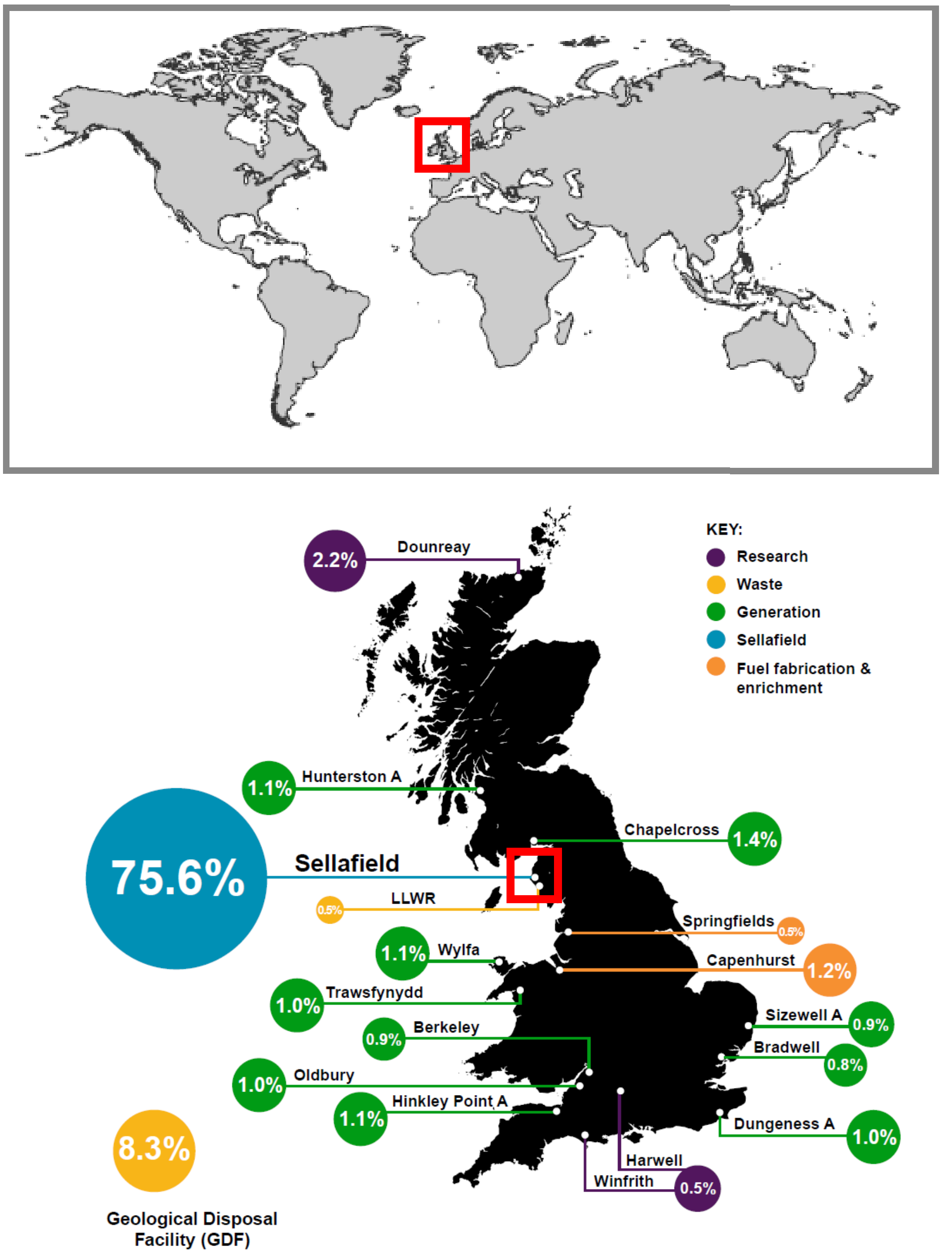


Figure 13 – Global and national location of Sellafield, UK. The bottom figure is adapted and redrawn from the original, available on the UK Government's website,[96] where circles represent total contribution to UK nuclear remediation clean-up costs.

The subsurface of the Sellafield site is primarily superficial glacial deposits overlaying sandstone bedrock.[97] Groundwater flow away from the Sellafield site tends towards the Irish Sea where, along with authorised discharges, contamination can become entrained in surrounding sediment. One such example is the Ravenglass estuary which contains ^{60}Co, which is amenable to EKR.[98] A schematic cross section of the Sellafield site is given in Figure 14.

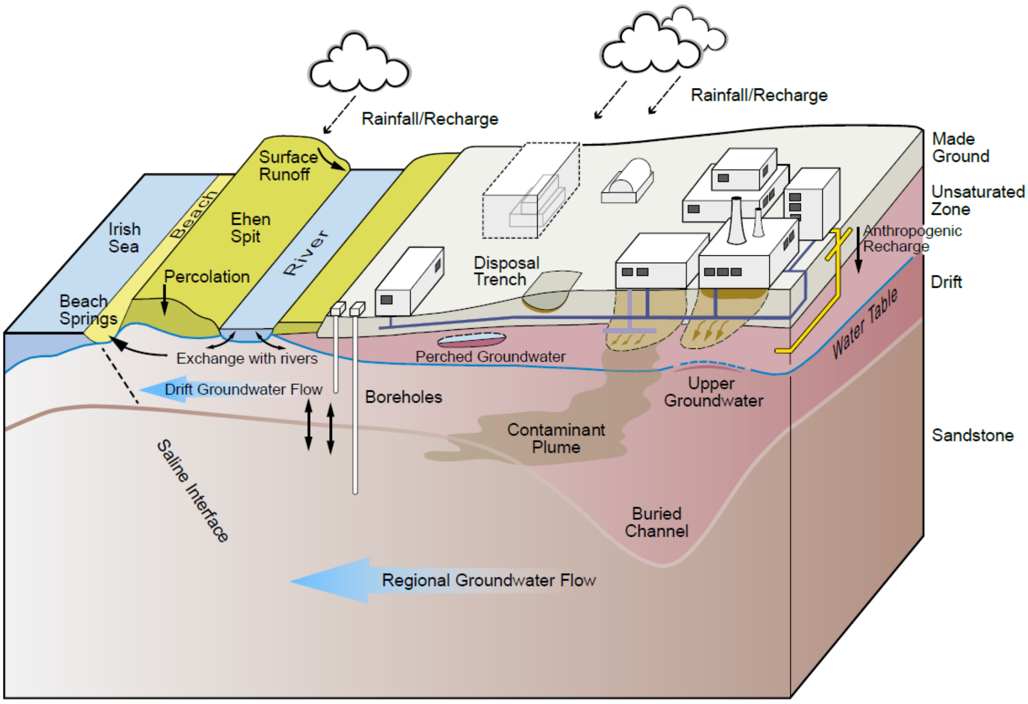


Figure 14 – Schematic of the ground conditions and local surface and subsurface water flows at the Sellafield site. Adapted and redrawn from the 2016 review of groundwater monitoring at Sellafield.[91]

The extensive contamination at Sellafield has driven numerous detailed investigations of EKR at the Sellafield site, led by the UK National Nuclear Laboratory (NNL). This includes its predecessors, British Nuclear Fuels Ltd. (BNFL) and Nexia Solutions Ltd. Due to the sensitive nature of these reports their results were not publicly disseminated, enabling us to present this work here for the first time.

The first example of EKR on the Sellafield site we are aware of was undertaken in the late 1990s by BNFL on three authentic Sellafield soils artificially spiked and saturated overnight with ^{137}Cs (caesium nitrate), ^{90}Sr (strontium nitrate) and transuranic alpha sources (including ^{239}Pu; 8 kBq.mL^{-1} solution of ^{239}Pu in 6 M nitric acid).[99] Here, ‘sample 1’ describes a spiked, clay-rich soil under static conditions, and ‘sample 2’ describes a spiked, sand-rich soil under flow of water; both samples were collected from points on the Sellafield site. EKR was assessed using a set up described in Figure 15, with radionuclide concentrations measured by scintillation after 14 days.

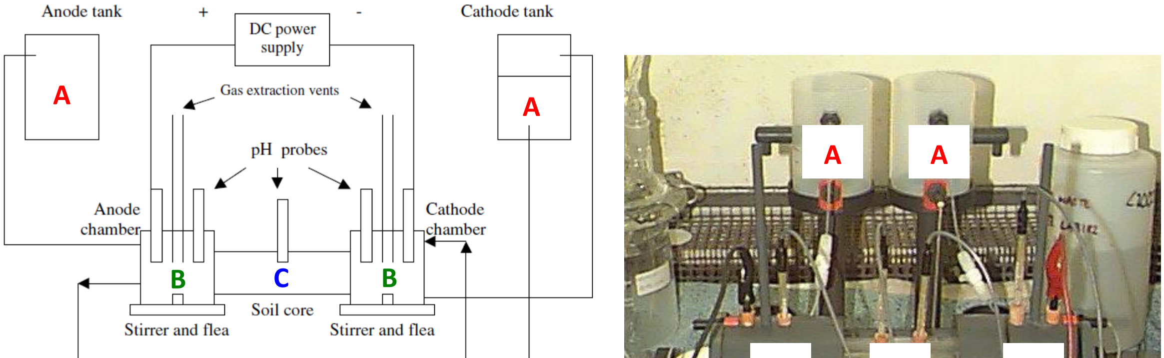


Figure 15 – Left, schematic of the EK cell used, and right, empty cell set-up utilised for the EKR analysis. The soil core (EKR chamber) was 7 cm long with an unspecified diameter. The electrolyte tanks (**A**), electrode chambers (**B**) and soil core, where the EK cell was housed (**C**), are highlighted.[99]

For both soil tests, 7 cm cores were selected and *ca*. 260 g material added to the soil core. The soil samples (samples 1 and 2) were treated for 14 days at 8 V with graphite electrodes. Under these conditions $^{137}\mathrm{Cs}^{+}$ or $^{90}\mathrm{Sr}^{2+}$ were significantly redistributed across the length of the soil sample. For the clayey spiked soil under static conditions, sample 1, the concentrations of $^{137}\mathrm{Cs}$ and $^{90}\mathrm{Sr}$ nearest the cathode decreased by 69 and 93%, respectively. When the stimulated effect of groundwater flow was added, sample 2, the redistribution was less effective, with reductions of 44% of $^{137}\mathrm{Cs}$ and 50% for $^{90}\mathrm{Sr}$ measured nearest the cathodes. Large errors in the α-spectroscopy measurements prevented an accurate assessment of $^{239}\mathrm{Pu}$ redistribution in the cores. A modified Tessier scheme was employed for sequential leaching, showing $^{137}\mathrm{Cs}$ and $^{90}\mathrm{Sr}$ were predominantly in exchangeable form (calcium chloride leach) and that $^{239}\mathrm{Pu}$ was predominantly leached with Fe and Mn oxides (hydroxylamine hydrochloride leach). Soil pH was also monitored, with the soil core and cathodic chamber becoming very basic (pH 12) after 14 days, and anodic chamber acidic (pH *ca*. 1.5) after the 14 day run.[99]

This set-up (Figure 15) was later used to assess EKR in a $^{90}\mathrm{Sr}$-, $^{239}\mathrm{Pu}$- and $^{126}\mathrm{Sn}$-contaminated, brucite ($\mathrm{Mg(OH)_2}$)-rich soil, representative of that present in a real leak area beneath a Sellafield waste storage facility.[100] Although the authors suggested EKR was not feasible on the Sellafield site (due to high alkaline buffering of the soil), we consider this to be an overly pessimistic assessment as the release of large quantities of alkaline material is not realistic of the entire site (on which carbonate concentrations can vary considerably).[101] The effectiveness of EKR at many areas of the Sellafield site with lower soil buffering capacity remains poorly researched.

Concerns over scalability (*e.g.* the 7 cm cell shown in Figure 15) were addressed separately on moving to a larger 40 L cell (Figure 16), where EKR on sandy or clayey Sellafield soils contaminated with $^{90}\mathrm{Sr}$ in simulant groundwater were examined.[102]

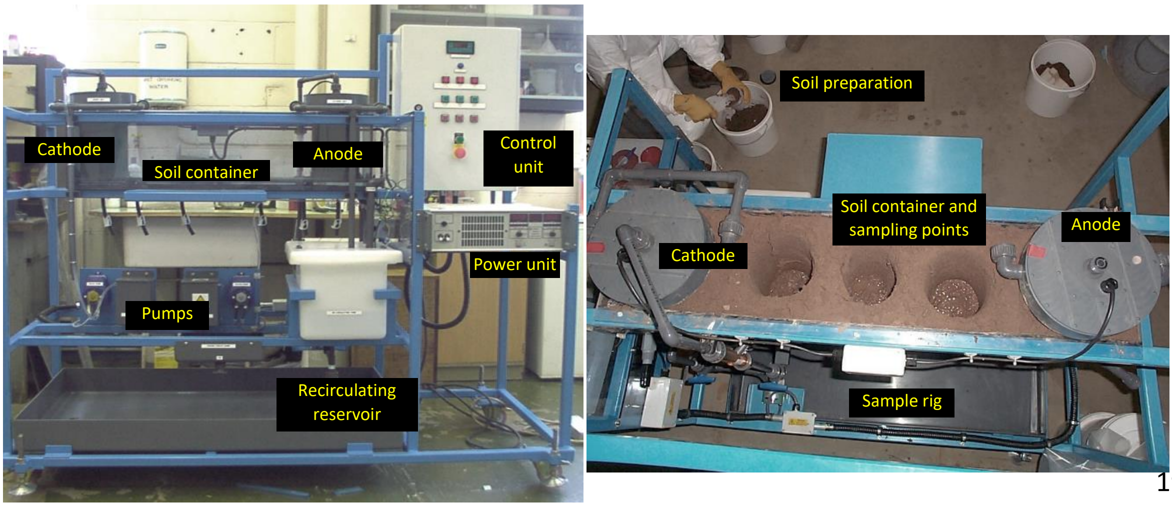


Figure 16 – Left, perspective view of the test rig used (LWH = 0.3 m x 0.75 m x 1.7 m) showing the pumping and electrical and control units, and right, birds-eye view of the test rig showing a full sample container, sample points and preparation of the soil.[102]

Measured β-activity values of 0.830 ± 0.254 Bq.g^{-1} (sandy soil) and 0.871 ± 0.251 Bq.g^{-1} (clayey soil) were obtained for both soils in this pumped system. As both soils were initially contaminated with ^{90}Sr at dangerous levels, the measured β-activities suggest that EKR was effective in reducing the contamination of these systems substantially.[102]

3.3.1 Ferric Iron Remediation and Stabilisation, FIRS

Building on earlier work by Hopkinson and Cundy *et al*.[98] scaled tests were also run to examine the potential application of EK processes to remotely generate *in-situ* iron barriers in simulated Sellafield soils and near-surface materials, Figure 17. This work utilises the *in-situ* generation of reactive iron barriers in soil when EKR is coupled with use of sacrificial steel electrodes.

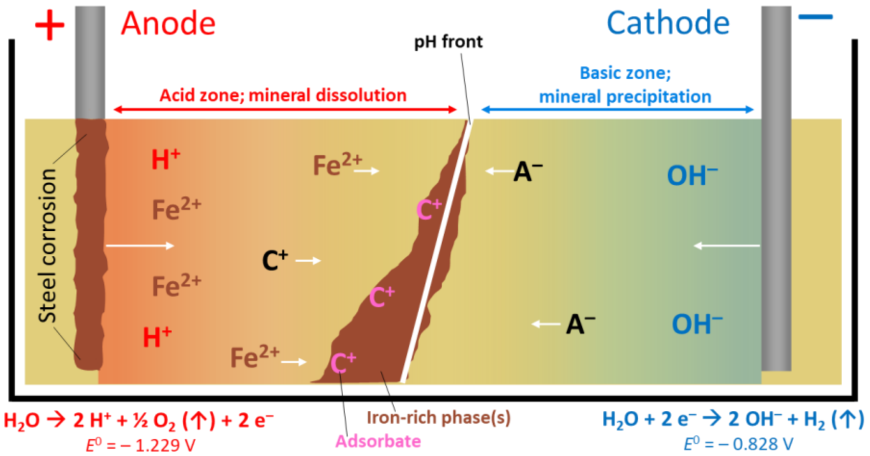


Figure 17 – A simplified EKR cell showing development of an iron-rich band in the FIRS technique and movement of ions. Cation (C^+) and anion (A^-) movement with pH gradient, towards electrodes of opposing charge, is shown. Water electrolysis half-cell values are *vs*. SHE.

Experiments operated up to metre-scales demonstrated the effective growth of iron rich barriers under site-relevant (ground)water salinities, at low voltages (≤ 0.5 V.cm^{-1}) over multiple months. These *in-situ* generated iron-rich "pans" may have significant use in providing reactive, *in-situ* barriers for groundwater flow control or groundwater remediation in higher permeability subsurface materials on the Sellafield site. We are developing these systems in collaboration with the UK nuclear industry and will report further in due course.

4. Towards Sustainable, Integrated EKR Techniques for Nuclear Sites

All EKR experiments discussed for these three sites are laboratory- or pilot-scale experiments, meaning that for nuclear sites, the scalability of EKR remains a pressing issue. While non-nuclear electrochemical remediation technologies are becoming commercially feasible,[18] evaluating the factors that influence the applicability of EKR is complex.

4.1 Key properties of EKR for use at nuclear sites

How, for example, does cost – the need to optimise public expenditure – balance with effecting the Best Practicable Environmental Option?[103] To answer this we have assessed previous work on the scalability of EKR,[16-21] and have identified four key areas (Figure 18) that influence the effectiveness of EKR with particular relevance for nuclear sites. These include i) properties of the substrate (soil, concrete, *etc.*) ii) properties of the contaminant, iii) electrical inputs and electrodes, and iv) cost and sustainability. The assessment presented here builds on a previous assessment made by the predecessor organisation to the NDA, Nexia Solutions Ltd.[104]

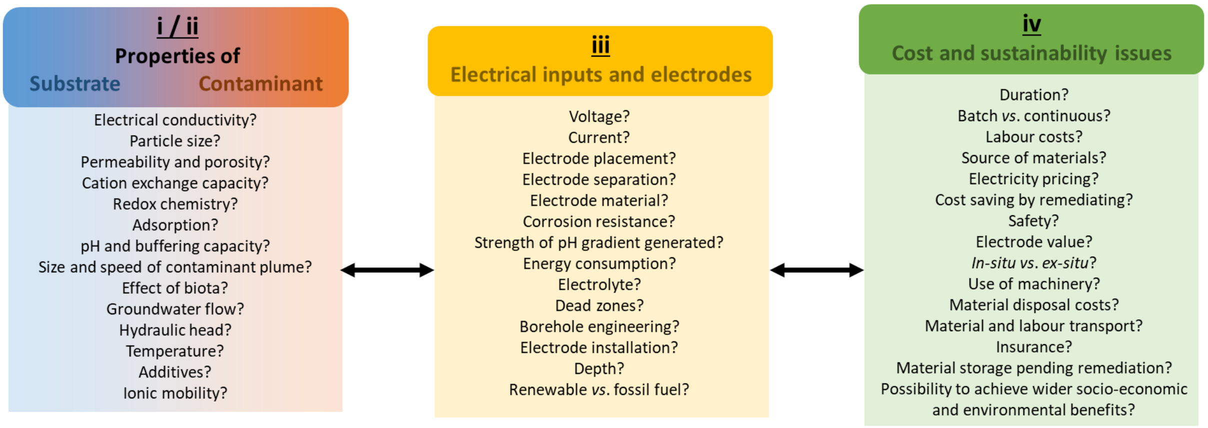


Figure 18 – Factors affecting the potential effectiveness of EKR at a specific site. Assessing these individually and understanding how all these issues affect others is key to understanding outcomes of EKR schemes.

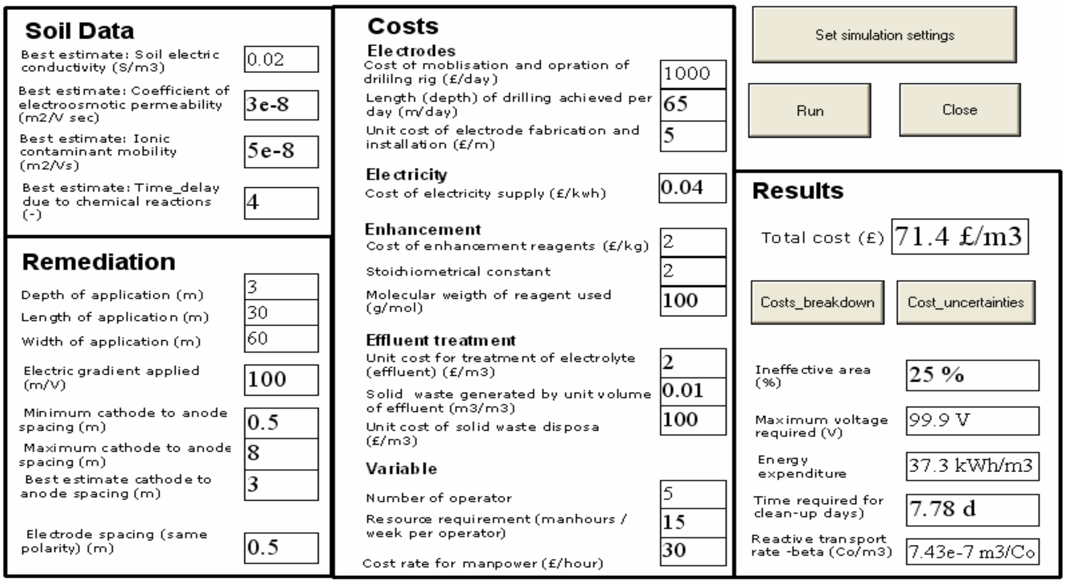
Firstly, consider the properties of the substrate to be remediated (Figure 18, i and ii). Soils, concretes, and sludges, *etc*. each have differing electrical conductivities, moisture levels, particle size distributions, cation exchange capacities, porosities and rheologies. Temperature and precipitation, groundwater flow and topologies will all also affect flow rates of a contaminant through a substrate. While the properties of a substrate are almost universally unique to a particular site, factors such as cation exchange capacities may be generalised dependent on soil type. In particular, clay-rich soils such as illite, montmorillonite, and kaolinite typically exhibit high acid/base buffering capacities, meaning strongly acidic conditions may be required to effectively transport contaminants. The physicochemical properties of a contaminant are also important (Figure 18, ii), and can include contaminant speciation (estimated using a Pourbaix phase diagram)[105] redox and pH sensitivity (*e.g.* Tc^{IV} *vs*. Tc^{VII}), adsorption behaviour (kinetics, Langmuir *vs*. Freundlich isotherms), and the size of a contaminant plume. The chemical form of contaminants is especially important as only dissolved solutes or colloids are amenable to EKR. It may also be necessary to inject additional reagents including weak acids, chelating agents or high ionic strength electrolytes, to enhance contaminant mobility.[106]

The electrical and electrode properties of the EKR unit (Figure 18, iii), such as voltage and current, direct flow of the contaminants to points in the cell; the higher the voltage, the faster the remediation. This must be balanced with electrode corrosion with acid-resistant materials at the anode, with platinum, graphite or coated titanium preferred as these are (typically) inert to dissolution. Any base-resistant and electrically-conducting material is generally acceptable as a cathode. In the case of sacrificial iron electrodes, electrode dissolution may actually be beneficial (Section 3.3.1). Expensive or valuable electrodes may be vulnerable to theft and thus require special security, although on tightly regulated nuclear sites this is unlikely to be a problem. Machinable electrodes, particularly hollow or porous ones, are beneficial as they can be combined with flushing technologies or act as PRBs in which the electrodes both migrate and trap selected contaminants. The electrochemical degradation of trichloroethylene has been achieved using machined porous graphite electrodes.[107]

A choice of vertical or horizontal electrode placement can be used to further control contaminant movement. Most experiments described here occur in only one or two-dimensions, with depth not considered. Clearly, in multi–centimetre or –metre deep soils, contaminant plume depth is important and further work is needed to establish the effect of electrode depth on contaminant remediation through a stratified soil profile. As discussed in Section 3.1, rather than actively remediating contaminated soil, it may be possible (through EKF) to passively prevent contamination in the first instance. This may be achieved by driving horizontal (sheet) electrodes into a soil but this requires carefully machined sheets of a pliable material, which are typically brittle and susceptible to fracturing if not handled carefully. Simple lines of electrode rods could alternatively be installed and, if buried to a certain depth, be used to generate horizontal fencing bands *in-situ*.[87] Although this is not a new idea[63] we are sceptical if the challenging engineering required to apply this on-site is commercially attractive compared to more established technologies (*e.g.* injection grouting). Avoiding electrical 'dead zones' (where field strength rapidly diminishes, meaning EKR ceases to be effective) is also important, and although changing electrode placement (*e.g.* hexagonal *vs*. circular *vs*. linear electrode arrays) has been examined[108, 109] it remains untested at scale. Large electrode spacings reduce the borehole and installation costs but increase the time taken to remediate a large area.

Finally, there are cost issues (Figure 18, iv) to be considered. The costs for EKR are low, rarely costing more than tens or maybe hundreds of USD for electricity or electrode and wiring materials.[70] Graphite or steel electrodes and wiring are available from home improvement stores, or *via* the Internet.[NOTE 3] Installation, labor and safety costs are the largest outlays, with borehole engineering and maintenance requiring trained operators over weeks to months. We estimate costs here to run to thousands of USD as salaries, safety, training, insurance and other costs are accounted for. Machinery, its rental or ownership costs and fuel may also need to be considered, depending on site-specific considerations.

Therefore, in deciding which variables (electrode placement, additives, *etc*.) to control (to maximise EKR effectiveness), the assessor is confronted with a complex, interacting set of parameters. In order to simplify the complex nature of EKR parameters, the NNL developed modelling software, Figure 19, within the GoldSim (Monte Carlo) package[NOTE 4] that assessed an EKR proposal on a number of technical and cost input metrics. These could be compared against output for similar models developed for other remedial techniques in order to support decisions on remedial approach, based on cost (rather than sustainability, below).



Soil Data

Best estimate: Soil electric conductivity (S/m3)	0.02
Best estimate: Coefficient of electroosmotic permeability (m2/V sec)	3e-8
Best estimate: Ionic contaminant mobility (m2/Vs)	5e-8
Best estimate: Time_delay due to chemical reactions (-)	4

Remediation

Depth of application (m)	3
Length of application (m)	30
Width of application (m)	60
Electric gradient applied (m/V)	100
Minimum cathode to anode spacing (m)	0.5
Maximum cathode to anode spacing (m)	8
Best estimate cathode to anode spacing (m)	3
Electrode spacing (same polarity) (m)	0.5

Costs

Electrodes	
Cost ot moblisation and opration of drililng rig (£/day)	1000
Length (depth) of drilling achieved per day (m/day)	65
Unit cost of electrode fabrication and installation (£/m)	5
Electricity	
Cost of electricity supply (£/kwh)	0.04
Enhancement	
Cost of enhancement reagents (£/kg)	2
Stoichiometrical constant	2
Molecular weigth of reagent used (g/mol)	100
Effluent treatment	
Unit cost for treatment of electrolyte (effluent) (£/m3)	2
Solid waste generated by unit volume of effluent (m3/m3)	0.01
Unit cost of solid waste disposa (£/m3)	100
Variable	
Number of operator	5
Resource requirement (manhours / week per operator)	15
Cost rate for manpower (£/hour)	30

Results

Total cost (£)	71.4 £/m3
Ineffective area (%)	25 %
Maximum voltage required (V)	99.9 V
Energy expenditure	37.3 kWh/m3
Time required for clean-up days)	7.78 d
Reactive transport rate -beta (Co/m3)	7.43e-7 m3/Co

Figure 19 – Example calculation being performed in the EKR cost simulator in the GoldSim package.

We are currently building on this GoldSIM model to develop our own tools to support the options appraisals for nuclear site decommissioning, and we discuss this further in Section 5.

4.1.1 EKR and Sustainability

Within the framework of increasing international standards over sustainable land remediation[110] (*e.g.* ISO18504:2017[111] and ASTM E2893[112]), key questions remain around the sustainability of EKR. Although cheap, the power and material requirements for EKR over prolonged periods of time can be resource intensive. To address this, EKR power inputs could be coupled to battery technology with renewable inputs (*e.g.* solar PV[113] and wind). Abundant electrode materials (*e.g.* steel rebar) may also help reduce the sustainability footprint of the EKR process.[51]

Combining EKR with other technologies (Section 4.2) could enhance the effectiveness of EKR and realise further sustainability benefits from the remediation process. For example, advantages to combining EKR with bio- or phytoremediation could include CO_2 sequestration, reduced dust emission, reduced material and resources cost, improved soil function, reduced waste generation, improved project lifespan and biodiversity benefits.[114] More generally the sustainability assessment criteria produced by the SuRF-UK (Sustainable Remediation Forum-UK), published in 2011[115] and updated in 2020,[116] provides a clear framework against which the sustainability of remediation options for contaminated land can be assessed.[117] Gill *et al*. have used the SuRF-UK framework to compare the sustainability benefits of EKR-Bio,[118] compared with alternative remediation options for a petroleum release site. The authors noted the strong performance of EKR-Bio compared to other intensive treatments, however, benefits are likely site-specific and methods to improve the sustainability of the EK-Bio treatment design (such as use of solar cells) had only a minor effect at the selected site.

4.2 Can other technologies help?

A significant advantage of EKR is that it can easily be applied in combination with other techniques. Our intention here is not to review the individual technologies but comment on how these have been or can be applied on nuclear sites with EKR.

4.2.1 EKR-Phyto and EKR-Bio

An recent contribution from Li *et al*. combines phyto-remediation with EKR for the remediation of uranium-contaminated soils using sunflower (*Helianthus*) and mustard (*Brassicaceae*) plants.[119] Although EKR-Phyto is known for non-nuclear remediation, for example in heavy metal (and Cs) remediation in paddy fields,[120] or petroleum-contaminated[121] land its use remains rare in radionuclide-contaminated soils.[122] In the study Li *et al*. spiked 0.1 g of uranium dioxide, uranium trioxide or uranyl nitrate per kilogram of Mississippi Delta soil, into which sunflowers and mustard plants were grown for 60 days. Phytoremediation without EKR after 60 days showed uranium removal efficiencies of only 0.8 – 4.3%. When this was repeated and combined with EKR after only 9 days, this increased to between 26 – 62% depending on the form of uranium (UO_2, UO_3 or uranyl nitrate) used. This increase is consistent with previous EKR-Phyto experiments using mustards in Cu-, Zn-, Cd- and Pb-contaminated paddy field soils, where similar removal efficiencies were reported (*ca*. 40 – 50%).[19] Although phytoremediation is slow (limited by plant lifecycles), combining it with EKR should offer significant benefits for the remediation of large areas of soil with low levels of shallow radionuclide contamination.[123]

We are not aware of any EKR-Bio schemes on nuclear sites, although EKR-Bio has received significant attention in recent years, including for the remediation of organics.[124] In particular, the group of Prof. E. K. Yanful (Western University, Canada) have undertaken numerous studies on the EKR-Bio of petroleum contaminated soils. Nuclear site wastes often contains organic contamination, including phenanthrene and other polyaromatic hydrocarbons (PAHs), phenols. At Sellafield, trichloroethylene, phthalates and phenols are noteworthy organic contaminants.[91] Important considerations for successful site-scale implementation of EKR-Bio for organic contaminants include prolonging the lifetime of indigenous microbiota, increasing metabolic rates and ensuring that metabolic pathways are not disrupted by the application of, or chemical by-products resulting from, the electrokinetic apparatus.[125] In particular, combining EKR-Bio reactors on the laboratory scale with solar (PV) cells was shown by Hassan *et al*. to operate effectively in the *mycobacterium*-based remediation of artificially contaminated kaolinite mixtures (2 mg of phenanthrene per gram of dry soil). One month of EKR in plastic cells (LWH 35x12x10 cm) with variable voltage resulted in removal of up to 50% of pre-experiment phenanthrene levels.[126] Further work on solar-powered EKR-Bio extended this to diesel fuel contaminated soil.[127] We are also aware of successful pilot-scale trials of EKR-Bio in low permeability soils contaminated with polychloroethylenes at a former industrial site in Denmark (*e.g.* Figure 20), showing that EKR-Bio is not limited to the laboratory-scale.

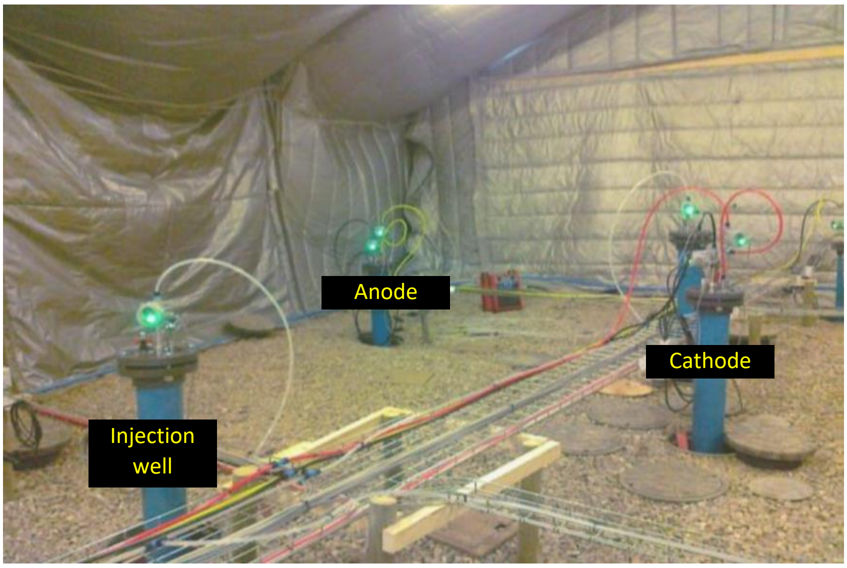


Figure 20 – Example of the EKR-Bio trial (2018) undertaken on tetrachloroethylene-contaminated soil in Denmark. Lactate and sodium hydroxide additives were used to stimulate bacterial growth.[128]

Here, EKR-Bio experiments produced three to five times as much ethene (a degradation product of polychloroethylenes) compared to bioremediation without EKR.[128]

4.2.2 EKR-ISCO (In-situ Chemical Oxidation)

In-situ chemical oxidation (ISCO) is a relatively well-established remediation technique where soluble oxidants are injected into a substrate to degrade (generally) organic pollutants. Similarly to EKR-Bio and EKR-Phyto, Section 4.2.1, recent developments have focused on ISCO of organic contaminants including chlorinated ethylenes and PAHs. Both are contaminants on active and legacy nuclear sites and recent work is of direct relevance to removing organic contaminants on nuclear sites. In particular, EKR enhances the transport of oxidants to desired points within an EKR cell by force-migrating ISCO reagents through low permeability soils.[129] In laboratory tests Chowdhury *et al*. demonstrated that permanganate ($Mn^{VII}O_4^{-}$) delivery was enhanced under EK through a silt-rich soil over a 41 day period, and reduced spiked trichloroethylene contamination more effectively than either EKR or ISCO alone.[130] Xu *et al*. have also shown that persulfate ($S_2O_8^{2-}$) delivery to PAH-contaminated soil is accelerated by the use of EK apparatus.[131] Although EKR-ISCO may be useful in low-permeability soils, the problem of residual oxidant contamination remains and as these (*e.g.* permanganate) are generally toxic,[132] their indiscriminate use is incompatible with a drive towards sustainable (*e.g.* mild) remediation technologies. We further note that redox-sensitive radionuclides (and metal contaminants) may not be amenable to EKR-ISCO as oxidation may alter the speciation of these contaminants.[133]

4.2.3 EKR-Nano, or EKR-Colloid, based technologies

As noted in section 3.3.1, sacrificial iron electrodes can be used to deliver Fe-rich phases into a soil, to reduce (*e.g.* Cr^{VI} to Cr^{III}, possibly also for Tc^{VII} to Tc^{IV}),[134] or sorb contaminants in the subsurface.[87, 98] There is also scope to electrophoretically “pump” or contain colloids and nanoparticles within an active treatment zone using the electric field, depending on their zeta potential. The movement of clays and colloids under an EK field is an area needing further research at nuclear sites. So-called ‘electro-grouting’ – where soils are stabilised through ion migration under an electric field – may also be combined with injection grouts to fill voids in building materials, again, electrophoretically.[135]

The electro-osmotic movement of calcium chloride and sodium silicates under an electric field is known to increase the shear strength of soft silty clays, and coupling EKR with nano-remediation[136] is effective for the degradation of organic pollutants.[137, 138] Engineered barrier repair or ground containment are other areas where EK may be combined with injection grouting,[139, 140] however we are not aware of any examples of EKR-Nano or electro-grouting applied on radioactively contaminated materials. We are examining combined approaches for this technology in our laboratories.

4.2.4 Electrokinetic Dewatering, EKD

Electrokinetic dewatering (EKD) is the electro-osmotic flow of water from anode to cathode, from where it can be removed. Although not strictly a combined technique EKD is a medium-to-high TRL technology with wide commercial applications, having been reported first in 1966[141] and applied for sludge thickening and dewatering for at least thirty years,[142] and thus we include it here. It is broadly applied at scale in various sectors including mining, water purification, and civil engineering.[143] This wide scalability makes it advantageous over other forms of ‘combined’ EKR techniques. It also aligns strongly with the waste minimisation objectives of the waste hierarchy. This was recently highlighted in work by Lamont-Black *et al*. on radioactively contaminated sludges on a working nuclear site.[144] A schematic of the setup used is given in Figure 21, alongside the material before and after processing, and the batch-scale setup employed.

Figure 21 – Top: batch scale processing of contaminated nuclear waste sludges on site; bottom: contaminated sludge before and after processing. Figures adapted from Lamont-Black *et al*.[144]

The sludges were dewatered in batches for 6 days at 30 V, with a total electricity consumption of 864 kWh. More than 400 m^3 of radioactively contaminated sludge was reduced to 30 m^3, a volume reduction of over 90%. Although this power consumption is high there is no reason why the EKD process could not be coupled with renewable energy sources to maximise energy efficiency. Given disposal costs of *ca*. \$52,000/£40,000 (UK, 2015) per tonne for contaminated waste, the cost benefit of EKD, if successfully incorporated into waste treatment workflows, may be significant. Sludges are among the most significant and problematic wasteforms on nuclear sites (at Sellafield, for example, almost 3000 m^3 of material is classed as 'sludge')[145] and EKD may offer another technique in the assessor's toolkit for remediating contaminated wastes.

4.3 TRL of EKR for Nuclear Site Applications

While many of the technologies discussed above are technologically feasible, few have been applied at scale, particularly as readily available, or "turn-key" solutions. This is particularly true for nuclear sites and partly reflects the considerable logistical challenges to applying these technologies at scale on what are (rightly) very heavily regulated industrial sites, rather than the technological inferiority of the EKR process itself. Where examples of scalable EKR processes are forthcoming, these are often *ex-situ* on individual batches of simulated materials, rather than *in-situ* under a continuous process on real site materials. This limits the current applicability of EKR and combined processes for use at nuclear sites, Table 3.

Table 3 – Our view on the current TRL status of EKR technologies for nuclear site application.

	TRL for nuclear site applications									
Technology	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**Summary Comments; nuclear *vs*. non-nuclear**
	Basic principles investigated	*Conceptualisation*	*Proof-of-concept*	*Laboratory experiment*	*Validate in real environment*	*Demonstrate in real environment*	*Operational prototype*	*Validate operational prototype*	*Fully operational system*	
EKR			🟧	🟨	🟩	🟩				**Nuclear 3 - 6:** On-site, *ex-situ*. Dependent on local geology. Drive towards sustainable remediation solutions increasing future use? **Non-nuclear 6 – 9+:** Widely demonstrated at scale.
EKR-Bio EKR-Phyto			🟧	🟨	🟩					**Nuclear 3 – 5:** Limited testing on nuclear sites; little work on radionuclides. Sustainability benefits unproven. **Non-nuclear 6 - 7:** EKR-Bio/-Phyto proven for heavy metals, organic, pesticides. Demonstrated in real systems (Section 4.2.1).
EKR-ISCO		🟥	🟧							**Nuclear 2 – 3:** Currently lab scale. Oxidants often toxic and unsuitable for redox-sensitive radionuclides. **Non-nuclear 4 - 6:** Technology demonstrated but at limited scale. Organic pollutants most amenable.
EKR-nano, or EKR-colloid, based systems			🟧	🟨	🟩	🟩				**Nuclear 3 – 6:** Many options; electro-grouting, FIRS, *etc*. Metre-scale known but not widely applied for radioactives. Site specific. **Non-nuclear 4 – 7:** Electro-grouting decades old and EKR-Nano becoming popular but neither is widely applied.
EKD					🟩	🟩	🟦			**Nuclear 6 – 7:** Site specific. Large scale commercial tests successful but only at Sellafield. **Non-nuclear 8 – 9+:** Widely applied at scale (Section 4.2.4).

We assess EKR for nuclear sites as between TRL 3 and 6; that is, demonstrated at the laboratory scale with limited intermediate and pilot scale work. For non-nuclear this extends to TRL 6 – 9+, as electrokinetics are widely applied for construction and mining applications.[146] There are significant practical issues limiting the wider scale applicability of EKF and EKR-ISCO for nuclear sites (for EKR-ISCO, organic pollutants in non-nuclear applications are most applicable), although combining EKR with other approaches (*e.g.* electro-grouting or EKR-Nano) is an expanding research area with growing future scope to address on-site contamination, sustainably.

5. Concluding Remarks and Scaling up

To answer the first of the three questions posed in the introduction, there has been considerable progress in EKR over the last decade. Since the last major review in 2009, EKR has been studied on new wasteforms and elements, including $^{134/137}$Cs-contaminated ash, uranium-contaminated concretes, gravels, ^{60}Co/^{137}Cs/$^{152/154}$Eu-contaminated spent graphite, and ^{99}Tc contaminated Mexican soil. Where reported, remediation efficiencies are high (often 90%+). Based on our assessment in Table 3, Figure 22 indicates elements we believe are amenable to EKR on nuclear sites. Diagonal shading indicates elements that have radionuclides of interest, but on which EKR has not been performed, or pre-2009. These include calcium (chemically analogous to strontium), nickel (mobile in experiments on naturally occurring Ni)[147], tin (non-nuclear mining waste, pre-2009),[148] iodine (Hg-selective EKR lixivant)[149] and radium and thorium (pre-2009 removal from clayey soils).[150]

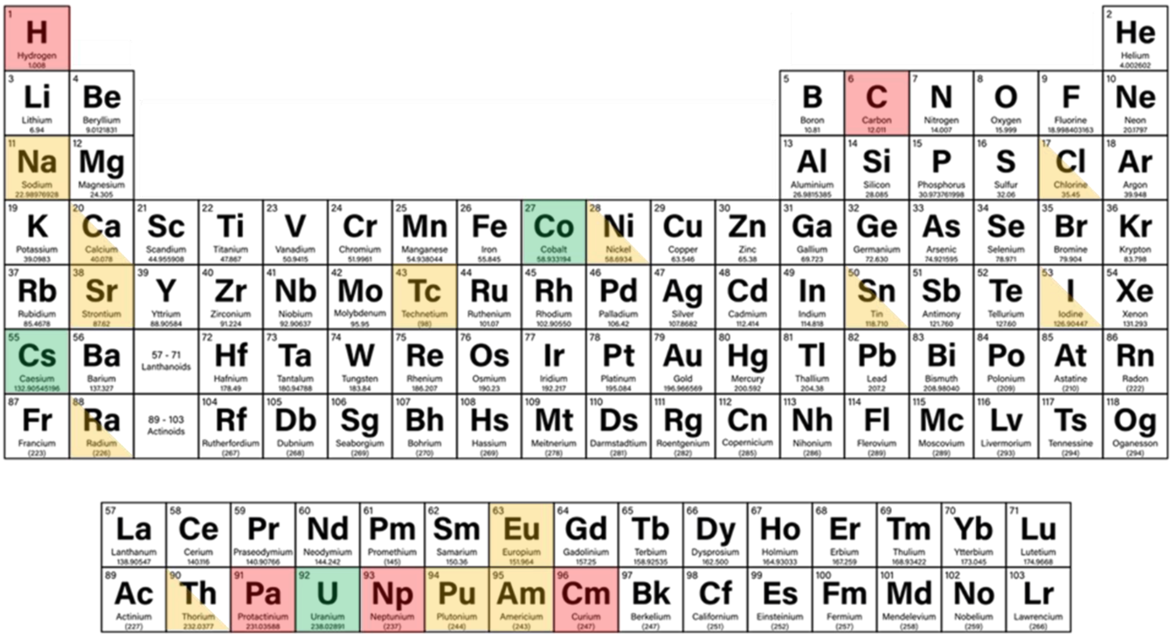


Figure 22 – Periodic table of elements with common isotopes present in nuclear sites applicable to EKR (Tables 1 and 2). Elements in red require further testing; yellow, limited work in past 10 years; diagonal yellow, demonstrated but not on a radionuclide and/or more than 10 years ago; and green, multiple studies showing EKR is useful in mobilising these elements. Elements with isotopes of half-lives < 1 year are excluded.

By moving from the laboratory to intermediate and even pilot scale, a number of authors have shown that even in tonnes of contaminated material removal efficiencies remain high (*ca*. 80%+), competitive with other technologies. Although these studies are *ex-situ* and employ washing (soils) or ball milling (concretes, gravels), they are the first step in demonstrating the wider feasibility of EKR for problem nuclides (*e.g.* through on-site, *ex-situ* applications).

Further, many of the radionuclides we consider in Figure 22 have traditionally been overlooked, because detecting low energy alpha or beta emissions from at least one of their isotopes is challenging. These include ^{3}H (Section 3.3), ^{14}C, ^{41}Ca, ^{63}Ni, ^{90}Sr, ^{99}Tc, ^{129}I, $^{239-241}Pu$, and ^{244}Cm, *via* non-destructive techniques.[151] Although some advances have been made with these DTM radionuclides since 2009 (Section 2.3),[51, 55, 152, 153] there remains a lack of research examining the remediation of these DTMs at scale, particularly EKR. This is despite being key contaminants in common materials found at nuclear sites, such as graphite, concrete, steel, water and soil.[154] Work to expand EKR for the treatment of DTM radionuclides is currently being undertaken in our laboratories with industrial partners and will be reported in due course.

For answers to the two remaining questions (EKR at selected nuclear sites, and further application of EKR), scalability remains problematic. Examples of EKR at the site scale (Hanford, Fukushima) do not yet exist and those at the pilot scale (Sellafield) are limited by the small number of studies in real site conditions. Off-the-shelf or “turn-key” EKR systems or solutions have not been applied, and given the large number of factors that can influence the success, duration and cost of EKR schemes (Figure 18), this is not surprising. However, the most important factors are likely to include the soil type, groundwater flow, water table depth, primary (radionuclide) contaminant, presence or absence of co-contaminants, required duration, and ionic mobility of contaminants. As these vary from site to site, we suggest that site-specific modelling of EKR rather than a general approach will be most effective in scaling EKR technologies up. Effective communication with stakeholders is vital to ensure knowledge is transferred between academic and industrial stakeholders most effectively.[118, 155-157] Decision support tools (DSTs) are models designed to help achieve this, ensuring stakeholders have access to relevant information in a form convenient to them. Several DSTs over the last decade or so have been produced to supplement the (sustainable) remediation of contaminated land.[155, 156, 158] Through the TRANSCEND consortium we are developing EKR DSTs with a view towards site-scale application, and we will report on this in due course.

We also suggest that combining EKR with other *in-situ* technologies may offer enhanced benefits *versus* EKR alone. Options that we consider most advantageous include EKR-Bio and EKR-Phyto (long duration but high potential for wider sustainability benefits), EKR-nano (easily combined, *e.g.* FIRS, electro-grouting) and EKD (which has demonstrated commercial viability). There are significant barriers to practical application of large-scale EKF (e.g. high continuous power requirement) or EKR-ISCO (harmful oxidants, possibility of enhanced mobilisation of some radionuclides). In terms of sustainability, although a full sustainability assessment of EKR-Bio for use on petroleum sites was recently published,[118] such an assessment for use of EKR on nuclear sites is lacking, and urgently needed. This would provide assessors with the information they need to decide the best technology to remediate a given site, sustainably, reflecting the growth of sustainable remediation guidelines[159] and legislation.[160]

CRediT authorship contribution statement

Jamie M. Purkis: Conceptualization, Methodology, Investigation, Validation, Visualization, Writing – original draft, Writing – review & editing

Phil E. Warwick: Funding acquisition, Supervision, Writing – review & editing

James Graham: Software, Validation, Writing – review & editing

Shaun D. Hemming: Investigation, Writing – review & editing

Andy B. Cundy: Conceptualization, Methodology, Investigation, Project administration, Supervision, Validation, Writing – original draft, Writing – review & editing, Funding acquisition

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Notes

Note 1: The UK Government-funded TRANSCEND consortium (Transformative Science and Engineering for Nuclear Decommissioning, https://transcendconsortium.org/) is a £9.4 million (*ca*. \$12.2 million) research programme of 40 research projects across industry and academia to address some of the key challenges remaining in nuclear decommissioning and waste management.

Note 2: The "ice-wall" has been widely reported in international news and TEPCO documents. TEPCO retains updated information on the status of the scheme, see https://www.tepco.co.jp/en/decommision/planaction/landwardwall/index-e.html. International news agencies carrying the stories of interest regarding the status of the "ice-wall" scheme include Reuters (https://www.nytimes.com/2016/08/30/science/fukushima-daiichi-nuclear-plant-cleanup-ice-wall.html), the New York Times (https://www.nytimes.com/2016/08/30/science/fukushima-daiichi-nuclear-plant-cleanup-ice-wall.html) and the Japan Times (https://www.japantimes.co.jp/news/2016/07/20/national/first-tepco-admits-ice-wall-cant-stop-fukushima-no-1-groundwater/).

Note 3: For example, Walmart.com (here) and Amazon.co.uk (here) both stock common electrode materials. These are sold to the public free of restrictions with next-day delivery options available.

Note 4: The GoldSIM software package (https://www.goldsim.com/) is a probabilistic simulator that employs Monte Carlo methods used in the analysis of radioactive waste management. Here, the NNL and partners developed a bespoke simulator with pre-defined parameters such as operator cost, *etc*., to act as a DST when assessing the feasibility of EKR for a given site. The proprietary simulator remains the property of the NNL, NDA, and selected partners.

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References

1. *Technologies for Remediation of Radioactively Contaminated Sites*, Report IAEA-TECDOC-1086, IAEA, Austria, 1999.
2. *Technology screening tools for radioactively contaminated sites*, Report EPA 402-R-96-017, US Environmental Protection Agency (EPA), 1996.
3. *Technology Reference Guide for Radioactively Contaminated Media*, Report EPA - 402-R-07-004, US Environmental Protection Agency (EPA), 2007.
4. *Remediation of Sites with Dispersed Radioactive Contamination*, Report STI/DOC/010/424, IAEA, Vienna, Austria, 2004.
5. *Remediation Technologies for Radioactive Contaminated Land on Nuclear Licensed Sites*, Report NT/7225002059/R2043, TÜV SÜD – Nuclear Technologies Division and Office of Nuclear Regulation (ONR), UK, 2020.
6. B. Kornilovich, N. Mishchuk, K. Abbruzzese, G. Pshinko and R. LKlishchenko, Enhanced electrokinetic remediation of metals-contaminated clay, *Colloids Surf. A Physicochem. Eng. Asp.*, 2005, **265**, 114-123.

7. S. V. Ho, C. Athmer, P. W. Sheridan, B. M. Hughes, R. Orth, D. McKenzie, P. H. Brodsky, A. Shapiro, R. Thornton, J. Salvo, D. Schultz, R. Landis, R. Griffith and S. Shoemaker, The Lasagna Technology for In Situ Soil Remediation. 1. Small Field Test, *Environ. Sci. Technol.*, 1999, **33**, 1086-1091.

8. *Basic principles of radioactive waste management*, Office for Nuclear Regulation (ONR), the Environment Agency, the Scottish Environment Protection Agency and Natural Resources Wales, UK, 2015.

9. C. Cameselle and S. Gouveia, Electrokinetic remediation for the removal of organic contaminants in soils, *Curr. Opin. Electrochem.*, 2018, **11**, 41-47.

10. T. D. Pham and M. Sillanpää, Electrokinetic remediation of organic contamination, *Environ. Tech. Rev.*, 2015, **4**, 103-117.

11. A. Saini, D. N. Bekele, S. Chadalavada, C. Fang and R. Naidu, A review of electrokinetically enhanced bioremediation technologies for PHs, *J. Environ. Sci.*, 2020, **88**, 31-45.

12. K. R. Reddy and C. Cameselle, *Overview of Electrochemical Remediation Technologies*, in *Electrochemical Remediation Technologies for Polluted Soils, Sediments and Groundwater*, eds. K. R. Reddy and C. Cameselle, John Wiley & Sons, Inc., 2009, ch. 1, pp. 1-28.

13. A. T. Lima, A. Hofmann, D. Reynolds, C. J. Ptacek, P. V. Cappellen, L. M. Ottosen, S. Pamukcu, A. Alshawabekh, D. M. O'Carroll, C. Riis, E.Cox, D.B.Gent, R.Landis, J.Wang, A.I.A.Chowdhury, E.L.Secord and A.Sanchez-Hachair, Environmental Electrokinetics for a sustainable subsurface, *Chemosphere*, 2017, **181**, 122-133.

14. G. C. C. Yang, Integrated electrokinetic processes for the remediation of phthalate esters in river sediments: A mini-review, *Sci. Total Environ.*, 2019, **659**, 963-972.

15. Y. B. Acar, R. J. Gale, A. N. Alshawabkeh, R. E. Marks, S. Puppala, M. Bricka and R. Parker, Electrokinetic remediation: Basics and technology status, *J. Haz. Mater.*, 1995, **40**, 117-137.

16. R. López-Vizcaíno, V. Navarro, M. J. Léon, C. Risco, M. A. Rodrigo, C. Sáez and P. Canizares, Scale-up on electrokinetic remediation: Engineering and technological parameters, *J. Haz. Mater.*, 2016, **315**, 135-143.

17. A. A. Suied, S. A. A. Tajudin, M. N. Zakaria and A. Madun, Potential Electrokinetic Remediation Technologies of Laboratory Scale into Field Application- Methodology Overview, *J. Phys.: Conf. Ser.*, 2018, **995**, 012083.

18. E. Lacasa, S. Cotillas, C. Saez, J. Lobato, P. Canizares and M. A. Rodrigo, Environmental applications of electrochemical technology. What is needed to enable full-scale applications?, *Curr. Opin. Electrochem.*, 2019, **16**, 149-156.

19. A. T. Yeung and Y.-Y. Gu, A review on techniques to enhance electrochemical remediation of contaminated soils, *J. Haz. Mater.*, 2011, **195**, 11-29.

20. B.-K. Kim, K. Baek, S.-H. Jo and J.-W. Yang, Research and field experiences on electrokinetic remediation in South Korea, *Sep. Purif. Technol.*, 2011, **79**, 116-123.

21. A. T. Yeung, Milestone developments, myths, and future directions of electrokinetic remediation, *Sep. Purif. Technol.*, 2011, **79**, 124-132.

22. V. A. Korolev, *Electrokinetic Removal of Radionuclides*, in *Electrochemical Remediation Technologies for Polluted Soils, Sediments and Groundwater*, eds. K. R. Reddy and C. Cameselle, John Wiley & Sons, Inc., 2009, DOI: https://onlinelibrary.wiley.com/doi/10.1002/9780470523650.ch5, ch. 5, pp. 127-139.

23. D. E. Ellis and P. W. Hadley, Sustainable remediation white paper—Integrating sustainable principles, practices, and metrics into remediation projects, *Remediation*, 2009, **19**, 5-114.

24. Y. B. Acar and A. N. Alshawabkeh, Principles of electrokinetic remediation, *Environ. Sci. Technol.*, 1993, **27**, 2638-2647.

25. A. De Rose, M. Buna, C. Strazza, N. Olivieri, T. Stevens, L. Peeters and D. Tawil-Jamault, *Technology readiness level: Guidance principles for renewable energy technologies*, Report EUR 27988 EN, European Union (EU), Brussels, 2017.

26. S. C. Sheppard, L. H. Johnson, B. W. Goodwin, J. C. Tait, D. M. Wuschke and C. C. Davison, Chlorine-36 in nuclear waste disposal—1. Assessment results for used fuel with comparison to 129I and 14C, *Waste Manage.*, 1996, **16**, 607-614.

27. X. Hou, Radiochemical determination of 41 Ca in nuclear reactor concrete, *Radiochim. Acta*, 2005, **93**, 611.

28. C. C. Lin, *Radiochemistry in Nuclear Power Reactors*, Report NAS-NS-3119, National Research Council (NRC; USA), Pleasanton, California, 1996.

29. M. F. Buehler, J. E. Surma and J. W. Virden, *In situ soil remediation using electrokinetics (PNL-SA-24081)* presented in part at the Thirty-Third Hanford Symposium on Health and the Environment, Richland, Washington, USA, 1994.

30. K. S. Leonard, D. McCubbin and B. R. Harvey, Chemical speciation and environmental behaviour of 60Co discharged from a nuclear establishment, *J. Environ. Radioact.*, 1993, **20**, 1-21.

31. G.-N. Kim, B.-I. Yang, W.-K. Choi, K.-W. Lee and J.-H. Hyeon, Washing-electrokinetic decontamination for concrete contaminated with cobalt and cesium, *Nucl. Eng. Technol.*, 2009, **41**.

32. G.-N. Kim, W.-K. Choi and K.-W. Lee, Decontamination of radioactive concrete using electrokinetic technology, *J. Appl. Electrochem.*, 2010, **40**, 1209-1216.

33. A. J. Parker, M. J. Joyce and C. Boxall, Remediation of 137Cs contaminated concrete using electrokinetic phenomena and ionic salt washes in nuclear energy contexts, *J. Haz. Mater.*, 2017, **340**, 454-462.

34. K. R. Reddy, *Electrokinetic remediation of soils at complex contaminated sites: Technology status, challenges, and opportunities*, in *Coupled Phenomena in Environmental Geotechnics*, eds. M. Manassero, A. Dominijanni, S. Foti and G. Musso, CRC Press, London, 2013, pp. 131-147.

35. G.-N. Kim, B.-I. Yang, W.-K. Choi and K.-W. Lee, Development of vertical electrokinetic-flushing decontamination technology to remove 60Co and 137Cs from a Korean nuclear facility site, *Sep. Purif. Technol.*, 2009, **68**, 222-226.

36. G.-N. Kim, S.-S. Lee, D.-B. Shon, K.-W. Lee and U.-S. Chung, Development of pilot-scale electrokinetic remediation technology to remove 60Co and 137Cs from soil, *J. Ind. Eng. Chem.*, 2010, **16**, 986-991.

37. G.-N. Kim, S.-S. Kim, U.-R. Park and J.-K. Moon, Decontamination of Soil Contaminated with Cesium using Electrokinetic-electrodialytic Method, *Electrochim. Acta*, 2015, **181**, 233-237.

38. G.-N. Kim, S.-S. Kim, H.-M. Park, W.-S. Kim, U.-R. Park and J.-K. Moon, Cs-137 and Cs-134 removal from radioactive ash using washing–electrokinetic equipment, *Ann. Nucl. Energy*, 2013, **57**, 311-317.

39. W. Briner, The Toxicity of Depleted Uranium, *Int. J. Environ. Res. Public Health*, 2010, **7**, 303-313.

40. M. Gavrilescu, L. V. Pavel and I. Cretescu, Characterization and remediation of soils contaminated with uranium, *J. Haz. Mater.*, 2009, **163**, 475-510.

41. G.-N. Kim, D.-B. Shon, H.-M. Park, K.-W. Lee and U.-S. Chung, Development of pilot-scale electrokinetic remediation technology for uranium removal, *Sep. Purif. Technol.*, 2011, **80**, 67-72.

42. W.-S. Kim, S.-S. Kim, G.-N. Kim, U.-R. Park and J.-K. Moon, Improved Treatment Technique for the Reuse of Waste Solution Generated from a Electrokinetic Decontamination System, *J. Nucl. Fuel Cycle Waste Technol. (Korean)*, 2014, **12**, 1-6.

43. G.-N. Kim, D.-B. Shon, H.-M. Park, K.-H. Kim, K.-W. Lee and J.-K. Moon, Precipitation-Filtering Method for Reuse of Uranium Electrokinetic Leachate, *J. Nucl. Fuel Cycle Waste Technol. (Korean)*, 2011, **9**, 63-71.

44. G.-N. Kim, S.-S. Kim, J.-K. Moon and J.-H. Hyun, Removal of uranium from soil using full-sized washing electrokinetic separation equipment, *Ann. Nucl. Energy*, 2015, **81**, 188-195.

45. G.-N. Kim, I. Kim, S.-S. Kim and J.-W. Choi, Removal of uranium from contaminated soil using indoor electrokinetic decontamination, *J. Radioanal. Nucl. Chem.*, 2016, **309**, 1175-1181.
46. S.-S. Kim, G.-N. Kim, U.-R. Park and J.-K. Moon, Development of a practical decontamination procedure for uranium-contaminated concrete waste, *J. Radioanal. Nucl. Chem.*, 2014, **302**, 611-616.
47. S.-S. Kim, W.-S. Kim, G.-N. Kim, H.-M. Park, U.-R. Park and J.-K. Moon, Decontamination of uranium-contaminated concrete, *Journal of Radioanalytical and Nuclear Chemistry*, 2013, **298**, 973-980.
48. A. Y. Yurchenko, Y. V. Karlin, A. N. Nikolaev, O. K. Karlina and A. S. Barinov, Decontamination of radioactive concrete, *At. Energy*, 2009, **106**, 225-230.
49. U.-K. Park, G.-N. Kim, S.-S. Kim and J.-K. Moon, Decontamination of Uranium-Contaminated Gravel, *J. Nucl. Fuel Cycle Waste Technol. (Korean)*, 2015, **13**, 35-43.
50. G.-N. Kim, U.-R. Park, S.-s. Kim, W.-S. Kim, J.-K. Moon and J.-h. Hyun, Decontamination of gravels contaminated with uranium, *Ann. Nucl. Energy*, 2014, **72**, 367-372.
51. K. Agnew, A. B. Cundy, L. Hopkinson, I. W. Croudace, P. E. Warwick and P. Purdie, Electrokinetic remediation of plutonium-contaminated nuclear site wastes: Results from a pilot-scale on-site trial, *J. Haz. Mater.*, 2011, **186**, 1405-1414.
52. Radioactive Substances Act, 1993, UK Government
53. E. V. Bespala, M. V. Antonenko, D. O. Chubreev, A. V. Leonov, I. Y. Novoselov, A. P. Pavlenko and V. N. Kotov, Electrochemical treatment of irradiated nuclear graphite, *J. Nucl. Mater.*, 2019, **526**, 151759.
54. *World reference base for soil resources 2014: International soil classification system for naming soils and creating legends for soil maps (Update 2015)*, Food and Agricultural Organisation (FAO) of the United Nations (UN), Rome, 2015.
55. V. Valdovinos, F. Monroy-Guzmán and E. Bustos, Electrokinetic removal of radionuclides contained in scintillation liquids absorbed in soil type Phaeozem, *J. Environ. Radioact.*, 2016, **162-163**, 80-86.
56. T. Tasutaka and W. Naito, Assessing cost and effectiveness of radiation decontamination in Fukushima Prefecture, Japan, *J. Environ. Radioact.*, 2016, **151**, 512-520.
57. O. Evrard, J. P. Laceby and A. Nakao, Effectiveness of landscape decontamination following the Fukushima nuclear accident: a review, *SOIL*, 2019, **5**, 333-350.
58. M. Okumura, S. Kerisit, I. C. Bourg, L. N. Lammers, T. Ikeda, K. M. Rosso and M. Machida, Radiocesium interaction with clay minerals: Theory and simulation advances Post–Fukushima, *J. Environ. Radioact.*, 2018, **189**, 135-145.
59. R. Kikuchi, H. Mukai, C. Kuramata and T. Kogure, Cs–sorption in weathered biotite from Fukushima granitic soil, *J. Mineral. Petrol. Sci.*, 2015, **110**, 126-134.
60. K. Tanaka, N. Watanabe, S. Yamasaki, A. Sakaguchi, Q. Fan and Y. Takahashi, Mineralogical control of the size distribution of stable Cs and radiocesium in riverbed sediments, *Geochem. J.*, 2018, **52**, 173-185.
61. K. Tanaka, Y. Takahashi, A. Y. A. Sakaguchi, M. Umeo, S. Hayakawa, H. Tanida, T. Saito and Y. Kanai, Vertical profiles of Iodine-131 and Cesium-137 in soils in Fukushima Prefecture related to the Fukushima Daiichi Nuclear Power Station Accident, *Geochem. J.*, 2012, **46**, 73-76.
62. J. Lehto, R. Koivula, H. Leinonen, E. Tusa and R. Harjula, Removal of Radionuclides from Fukushima Daiichi Waste Effluents, *Sep. Purif. Rev.*, 2019, **48**, 122-142.
63. R. López-Vizcaíno, C. Risco, J. Isidro, S. Rodrigo, C. Saez, P. Cañizares, V. Navarro and M. A. Rodrigo, Scale-up of the electrokinetic fence technology for the removal of pesticides. Part II: Does size matter for removal of herbicides?, *Chemosphere*, 2017, **166**, 549-555.
64. R. Lageman, *Preliminary assessment of the application of an electrokinetic ring fence for the removal of radionuclides from groundwater at Fukushima Daiichi Nuclear Power Plant*, Lambda Consult (Netherlands), 2014.

65. R. Lageman, *Demonstration Project for Seawater Purification Technologies*, Lambda Consult (Netherlands), 2014.

66. T. Miura, M. Kabir, M. Suzuki, S. Nakajima and S. Mori, Cesium removal from contaminated soil by electrokinetic process considering for slope areas of mountains, *IEEJ Trans. A Fundam. Mater. (translated from Japanese: Denki Gakkai Ronbunshi. A)*, 2015, **135**, 725-726.

67. A. H. Gallardo and A. Marui, The aftermath of the Fukushima nuclear accident: Measures to contain groundwater contamination, *Sci. Total Environ.*, 2016, **547**, 261-268.

68. A. J. Fuller, S. Shaw, M. B. Ward, S. J. Haigh, J. F. W. Mosselmans, C. L. Peacock, S. Stackhouse, A. J. Dent, D. Trivedi and I. T. Burke, Caesium incorporation and retention in illite interlayers, *Appl. Clay Sci.*, 2015, **108**, 128-134.

69. J. Koarashi, S. Nishimura, M. Atarashi-Andoh, T. Matsunaga, T. Sato and S. Nagao, Radiocesium distribution in aggregate-size fractions of cropland and forest soils affected by the Fukushima nuclear accident, *Chemosphere*, 2018, **205**, 147-155.

70. J. M. Purkis, A. Tucknott, I. W. Croudace, P. E. Warwick and A. B. Cundy, *submitted to Appl. Geochem.*

71. D. Harbottle and T. Hunter, 2019, Electrokinetic Separation for Enhanced Decontamination of Soils and Groundwater Systems (grant EP/S032797/1), Engineering and Physical Sciences Research Council (EPRSC), UK

72. T. S. Peretyazhko, J. M. Zachara, R. K. Kukkadapu, S. M. Heald, I. V. Kutnyakov, C. T. Resch, B. W. Arey, C. M. Wang, L. Kovarik, J. L. Phillips and D. A. Moore, Pertechnetate (TcO4−) reduction by reactive ferrous iron forms in naturally anoxic, redox transition zone sediments from the Hanford Site, USA, *Geochim. Cosmochim. Acta*, 2012, **92**, 48-66.

73. N. P. Qafoku, J. M. Zachara, C. Liu, P. L. Gassman, O. S. Qafoku and S. C. Smith, Kinetic Desorption and Sorption of U(VI) during Reactive Transport in a Contaminated Hanford Sediment, *Environ. Sci. Technol.*, 2005, **39**, 3157-3165.

74. J. K. Fredrickson, J. M. Zachara, D. L. Balkwill, D. Kennedy, S.-m. W. Li, H. M. Kostandarithes, M. J. Daly, M. F. Romine and F. J. Brockman, Geomicrobiology of High-Level Nuclear Waste-Contaminated Vadose Sediments at the Hanford Site, Washington State, *Appl. Environ. Microbiol.*, 2004, **70**, 4230.

75. J. P. McKinley, C. J. Zeissler, J. M. Zachara, R. J. Serne, R. M. Lindstrom, H. T. Schaef and R. D. Orr, Distribution and Retention of 137Cs in Sediments at the Hanford Site, Washington, *Environ. Sci. Technol.*, 2001, **35**, 3433-3441.

76. J. A. Beavers, S. Chawla, K. Evans, B. C. Rollins, K. M. Sherer, N. Sridhar, K. D. Boomer, C. L. Girardot, T. Venetz and L. M. Stock, History of the First Double-Shell High-Level Nuclear Waste Tank at the Hanford Site, *Corrosion*, 2019, **75**, 292-301.

77. *2019 Hanford Lifecycle Scope, Schedule and Cost Report*, Report DOE/RL-2018-45, Department of Energy (DoE), US, 2019.

78. *Hanford Site Groundwater Monitoring and Performance Report for 2009*, Report DOE/RL-2010-11, Department of Energy (DoE), US, 2010.

79. *Nuclear Site Remediation and Restoration during Decommissioning of Nuclear Installations: A Report by the NEA Co-operative Programme on Decommissioning*, Report NEA No. 7192, Nuclear Energy Agency (NEA) of the Organisation for Economic Cooperation and Development (OECD), 2014.

80. V. R. Vermeul, J. E. Szecsody, B. G. Fritz, M. D. Williams, R. C. Moore and J. S. Fruchter, An Injectable Apatite Permeable Reactive Barrier for In Situ 90Sr Immobilization, *Groundwater Monit. Remed.*, 2014, **34**, 28-41.

81. *Optimization of Groundwater Pump and Treat Systems at Hanford*, Report No. 031102-027, Office of Environmental Management, Department of Energy (DoE), US, 2004.

82. M. E. Brines, S. Simmons and J. Morse, *Startup of the New 200 West Pump-and-Treat, Hanford Site, Richland, Washington* presented in part at the Waste Management (WM) Symposia 2013, Phoenix, Arizona, US, 2013.

83. *Hanford Site Excavating, Trenching and Shoring Procedure (HSETSP)*, Report DOE-0344 Revision 3A, Department of Energy (DoE), US, 2013.

84. R. S. Skeen, K. R. Roberson, D. J. Workman, J. N. Petersen and M. Shouche, *In-situ bioremediation of Hanford groundwater* presented in part at the 1992 Federal Environmental Restoration Conference, Vienna, Virgina, US, 1992.

85. *In Situ Bioremediation for the Hanford Carbon Tetrachloride Plume*, Office of Environmental Management, Department of Energy (DoE), US, 1999.

86. H. B. Jung, J.-S. Yang and W. Um, Bench-scale electrokinetic remediation for cesium-contaminated sediment at the Hanford Site, USA, *J. Radioanal. Nucl. Chem.*, 2015, **304**, 615-625.

87. D. W. S. Faulkner, L. Hopkinson and A. B. Cundy, Electrokinetic generation of reactive iron-rich barriers in wet sediments: implications for contaminated land management, *Mineral. Mag.*, 2005, **69**, 749-757.

88. S. Tanaka, Gypsum precipitation enhanced by electrokinetic method and porewater chemistry in compacted montmorillonite, *Appl. Clay Sci.*, 2018, **161**, 482-493.

89. G. T. Lee, H. M. Ro and S. M. Lee, Effects of triethyl phosphate and nitrate on electrokinetically enhanced biodegradation of diesel in low permeability soils, *Environ. Technol.*, 2007, **28**, 853-860.

90. *Context Plan (2017-2026): Explaining the context of Sellafield's operations*, Sellafield Ltd. (UK), 2017.

91. *Sellafield Groundwater Monitoring Annual Data Review 2016*, Report LQTD000758, Sellafield Ltd. (UK), 2016.

92. *Guidlines for Drinking-Water Quality, Fourth Edition*, World Health Organisation (WHO), 2011.

93. *The Nuclear Decommissioning Authority: progress with reducing risk at Sellafield*, National Audit Office (NAO), UK, 2018.

94. *Nuclear Decommissioning Authority: Managing risk reduction at Sellafield*, National Audit Office (NAO), UK, 2012.

95. *Nuclear Provision - explaining the cost of cleaning up Britain's nuclear legacy*, Nuclear Decommissioning Authority (NDA), UK, 2015.

96. Nuclear Decommissioning Authority, Nuclear Provision: the cost of cleaning up Britain’s historic nuclear sites, https://www.gov.uk/government/publications/nuclear-provision-explaining-the-cost-of-cleaning-up-britains-nuclear-legacy/nuclear-provision-explaining-the-cost-of-cleaning-up-britains-nuclear-legacy, (accessed 16th October, 2020).

97. N. T. Smith, J. Shreeve and O. Kuras, Multi-sensor core logging (MSCL) and X-ray computed tomography imaging of borehole core to aid 3D geological modelling of poorly exposed unconsolidated superficial sediments underlying complex industrial sites: An example from Sellafield nuclear site, UK, *J. Appl. Geophys.*, 2020, **178**, 104084.

98. A. B. Cundy and L. Hopkinson, Electrokinetic iron pan generation in unconsolidated sediments: implications for contaminated land remediation and soil engineering, *Appl. Geochem.*, 2005, **20**, 841-848.

99. A. B. Eilbeck and P. E. Learmonth, *An Experimental Investigation into the Application of Electrokinetic Remediation to Contaminated Sellafield Soils*, Report RDR 1717, British Nuclear Fuels Ltd. (BNFL), UK, 1999.

100. A. Eilbeck and K. Lorimer, *Electrokinetic Remediation Studies on Soil Contaminated with B38 Leak Simulant*, Report RAT 1307, British Nuclear Fuels Ltd. (BNFL), UK, 2001.

101. J. Graham, *MSSS GEMS Phase 2: Characterisation of Soils and Strontium Sorption Batch Testing*, Report NNL 13224, National Nuclear Laboratory, NNL, 2015.

102. J. R. Brydie and I. Beadle, *Electrokinetic Remediation of Sellafield Soils in Preparation for Field Scale Remediation Trials*, Report NSTS 4965, British Nuclear Fuels Ltd. (BNFL), UK, 2004.

103. T. O'Riordan, Best Practicable Environmental Option (BPEO): A Case-study in Partial Bureaucratic Adaptation, *Environ. Conserv.*, 1989, **16**, 113-122.

104. N. Parton, *Remediation Technology Demonstrators: availability of version 1 of the cost and technology models for soil washing, permeable reactive barriers, in-situ stabilisation and electrokinetics*, Report NDA 1648.4, Nexia Solutions (UK), 2007.

105. N. Takeno, *Atlas of Eh-pH diagrams, Intercomparison of thermodynamic databases, Geological Survey of Japan Open File Report No. 419*, National Institute of Advanced Industrial Science and Technology, Research Center for Deep Geological Environments, 2005.

106. R. A. Wuana and F. E. Okieimen, Heavy Metals in Contaminated Soils: A Review of Sources, Chemistry, Risks and Best Available Strategies for Remediation, *ISRN Ecology*, 2011, **2011**, 402647.

107. L. Rajic, R. Nazari, N. Fallahpour and A. N. Alshawabkeh, Electrochemical degradation of trichloroethylene in aqueous solution by bipolar graphite electrodes, *J. Environ. Chem. Eng.*, 2016, **4**, 197-202.

108. R. S. Putra, Development of electrokinetic remediation for caesium: A feasibility study of 2D electrode configuration system, *IOP Conf. Ser.: Mater. Sci. Eng.*, 2016, **107**, 012015.

109. A. N. Alshawabkeh, R. J. Gale, E. Ozsu-Acar and R. M. Bricka, Optimization of 2-D Electrode Configuration for Electrokinetic Remediation, *J. Soil Contamin.*, 1999, **8**, 617-635.

110. R. P. Bardos, A. B. Cundy, J. W. N. Smith and N. Harries, Sustainable remediation, *J. Environ. Manage.*, 2016, **184**, 1-3.

111. *ISO 18504:2017: Soil quality - Sustainable remediation*, International Standards Organisation (ISO), 2017.

112. *ASTM E2893 - 16e1: Standard Guide for Greener Cleanups*, ASTM International, 2016.

113. I. Hassan, E. Mohamedelhassan and E. K. Yanful, Solar powered electrokinetic remediation of Cu polluted soil using a novel anode configuration, *Electrochim. Acta*, 2015, **181**, 58-67.

114. A. A. Juwarkar, Microbe-Assisted Phytoremediation for Restoration of Biodiversity of Degraded Lands: A Sustainable Solution, *Proc. Natl. Acad. Sci. India B*, 2012, **82**, 313-318.

115. *Annex 1: The SuRF-UK Indicator Set for Sustainable Remediation Assessment*, Sustainable Remediation Forum (SuRF) and Contaminated Land: Application in Real Environments (CL:AIRE), UK, 2011.

116. *Supplementary Report 1 of the SuRF-UK Framework: A General Approach to Sustainability Assessment for Use in Acheiving Sustainable Remediation*, Sustainable Remediation Forum (SuRF) and Contaminated Land: Application in Real Environments (CL:AIRE), UK, 2020.

117. X. Li, P. Bardos, A. B. Cundy, M. K. Harder, K. J. Doick, J. Norrman, S. Williams and W. Chen, Using a conceptual site model for assessing the sustainability of brownfield regeneration for a soft reuse: A case study of Port Sunlight River Park (U.K.), *Sci. Total Environ.*, 2019, **652**, 810-821.

118. R. T. Gill, S. F. Thornton, M. J. Harbottle and J. W. N. Smith, Sustainability assessment of electrokinetic bioremediation compared with alternative remediation options for a petroleum release site, *J. Environ. Manage.*, 2016, **184**, 120-131.

119. J. Li, J. Zhang, S. L. Larson, J. H. Ballard, K. Guo, Z. Arslan, Y. Ma, C. A. Waggoner, J. R. White and F. X. Han, Electrokinetic-enhanced phytoremediation of uranium-contaminated soil using sunflower and Indian mustard, *Int. J. Phytoremed.*, 2019, **21**, 1197-1204.

120. X. Mao, F. X. Han, X. Shao, K. Guo, J. McComb, Z. Arslan and Z. Zhang, Electro-kinetic remediation coupled with phytoremediation to remove lead, arsenic and cesium from contaminated paddy soil, *Ecotoxicol. Environ. Saf.*, 2016, **125**, 16-24.

121. I. M. V. Rocha, K. N. O. Silva, D. R. Silva, C. A. Mártinez-Huitle and E. V. Santos, Coupling electrokinetic remediation with phytoremediation for depolluting soil with petroleum and the use of electrochemical technologies for treating the effluent generated, *Sep. Purif. Technol.*, 2019, **208**, 194-200.

122. H. Shahandeh and L. R. Hossner, Enhancement of uranium phytoaccumulation from contaminated soils, *Soil Sci.*, 2002, **167**, 269-280.

123. S. Chatterjee, A. Mitra, C. Walther and D. K. Gupta, *Plant Response Under Strontium and Phytoremediation*, in *Strontium Contamination in the Environment*, eds. P. Pathak and D. K. Gupta, Springer International Publishing, Cham, 2020, pp. 85-97.

124. R. T. Gill, M. J. Harbottle, J. W. N. Smith and S. F. Thornton, Electrokinetic-enhanced bioremediation of organic contaminants: A review of processes and environmental applications, *Chemosphere*, 2014, **107**, 31-42.

125. I. Hassan, E. Mohamedelhassan, E. Yanful and Z.-C. Yuan, *Enhancement of Bioremediation and Phytoremediation Using Electrokinetics*, in *Advances in Bioremediation and Phytoremediation*, ed. N. Shiomi, IntechOpen, 2018.

126. I. Hassan, E. Mohamedelhassan, E. K. Yanful and Z.-C. Yuan, Solar power enhancement of electrokinetic bioremediation of phenanthrene by Mycobacterium pallens, *Bioremediat. J.*, 2017, **21**, 53-70.

127. I. Hassan, E. Mohamedelhassan, E. K. Yanful and Z.-C. Yuan, *Solar powered electrokinetic bioremediation of diesel fuel contamination* presented in part at the International Symposium on Electrokinetic Remediation 2016 (EREM 2016), Abu Dhabi, United Arab Emirates (UAE), 2016.

128. C. Riis, M. Bymose, E. Cox, J. Wang, D. Gent and M. Terkelsen, *Successful pilot test of electrokinetic enhanced bioremediation (EK-BIO) as an innovative remedial approach for PCE/DNAPL source area* presented in part at the NORDROCS 2012: 4th Nordic Joint Meeting on Remediation of Contaminated Sites, Oslo, Norway, 2012.

129. M. Z. Wu, D. A. Reynolds, A. Fourie, D. Thomas and H. Prommer, *Field Implementation of Electrokinetic-ISCO Remediation* presented in part at the 2010 Fall Meeting of the American Geophysical Union, San Francisco, California, US, 2010.

130. A. I. A. Chowdhury, J. I. Gerhard, D. Reynolds, B. E. Sleep and D. M. O'Carroll, Electrokinetic-enhanced permanganate delivery and remediation of contaminated low permeability porous media, *Water Res.*, 2017, **113**, 215-222.

131. H. Xu, Y. Song, L. Cang and D. Zhou, Ion exchange membranes enhance the electrokinetic in situ chemical oxidation of PAH-contaminated soil, *J. Haz. Mater.*, 2020, **382**, 121042.

132. H. T. Kemp, R. G. Fuller and R. S. Davidson, Potassium Permanganate as an Algicide, *J. Am. Water Work. Assoc.*, 1966, **58**, 255-263.

133. M. Crimi, T. J. Simpkin, T. Palaia, B. G. Petri and R. L. Siegrist, *Systematic Approach for Site-Specific Engineering of ISCO*, in *In Situ Chemical Oxidation for Groundwater Remediation*, eds. R. L. Siegrist, M. Crimi and T. J. Simpkin, Springer New York, New York, NY, 2011, pp. 355-412.

134. L. Hopkinson, A. B. Cundy, D. W. S. Faulkner, A. Hansen and R. Pollock, *Electrokinetic stabilization of Cr(VI)-contaminated soils*, in *Electrochemical remediation technologies for polluted soils, sediments and groundwater*, eds. K. Reddy and C. Cameselle, John Wiley and Sons, Inc., New York, 2009, ch. 8, pp. 179-193.

135. A. N. Alshawabkeh and T. C. Sheahan, Soft soil stabilisation by ionic injection under electric fields, *Proc. Instit. Civil Eng. - Ground Improvement*, 2003, **7**, 177-185.

136. A. P. Ingle, A. B. Seabra, N. Duran and M. Rai, *Nanoremediation: A New and Emerging Technology for the Removal of Toxic Contaminants from Environment*, in *Microbial Biodegradation and Bioremediation*, ed. S. Das, Elsevier, Oxford, 2014, pp. 233-250.

137. H. I. Gomes, G. Fan, L. M. Ottosen, C. Dias-Ferreira and A. B. Ribeiro, *Nanoremediation Coupled to Electrokinetics for PCB Removal from Soil*, in *Electrokinetics Across Disciplines and Continents: New Strategies for Sustainable Development*, eds. A. B. Ribeiro, E. P. Mateus and N. Couto, Springer International Publishing, Cham, 2016, pp. 331-350.

138. H. I. Gomes, G. Fan, E. P. Mateus, C. Dias-Ferreira and A. B. Ribeiro, Assessment of combined electro–nanoremediation of molinate contaminated soil, *Sci. Total Environ.*, 2014, **493**, 178-184.

139. M. Bani Baker, M. Elektorowicz and A. Hanna, Electrokinetic nondestructive in-situ technique for rehabilitation of liners damaged by fuels, *J. Haz. Mater.*, 2018, **359**, 510-515.

140. C. Wong, M. Pedrotti, G. El Mountassir and R. J. Lunn, A study on the mechanical interaction between soil and colloidal silica gel for ground improvement, *Eng. Geol.*, 2018, **243**, 84-100.

141. L. G. Adamson, G. V. Chilingar, C. M. Beeson and R. A. Armstrong, Electrokinetic dewatering, consolidation and stabilization of soils, *Eng. Geol.*, 1966, **1**, 291-304.

142. J. G. Sunderland, Electrokinetic dewatering and thickening. I. Introduction and historical review of electrokinetic applications, *J. Appl. Electrochem.*, 1987, **17**, 889-898.

143. J. Colin, L.-B. John, H. David, A. David and G. Stephanie, Electrokinetic geosynthetics: from research to hype to practice, *Proc. Instit. Civil Eng. - Civil Engineering*, 2017, **170**, 127-134.

144. J. Lamont-Black, C. J. F. P. Jones and C. White, Electrokinetic geosynthetic dewatering of nuclear contaminated waste, *Geotext. Geomembr.*, 2015, **43**, 359-362.

145. *2016 UK Radioactive Waste & Materials Inventory: UK Radioactive Waste Inventory Report*, Department of Business, Energy and Industrial Strategy (BEIS) and Nuclear Decommissioning Authority (NDA), UK, 2017.

146. H. K. Hansen, L. M. Ottosen and A. B. Ribeiro, *Electrokinetic Soil Remediation: An Overview*, in *Electrokinetics Across Disciplines and Continents: New Strategies for Sustainable Development*, eds. A. B. Ribeiro, E. P. Mateus and N. Couto, Springer International Publishing, Cham, 2016, pp. 3-18.

147. M. Saleem, M. H. Chakrabarti, M. F. Irfan, S. A. Hajimolana, M. A. Hussain, B. H. Diya’uddeen and W. M. A. W. Daud, Electrokinetic Remediation of Nickel From Low Permeability Soil, *Int. J. Electrochem. Sci.*, 2011, **6**, 4264-4275.

148. C. C. Ho and K. C. Lee, Electrokinetic behaviour of tin tailings slurries in solutions of lanthanum and thorium salts, *Colloids Surf. A Physicochem. Eng. Asp.*, 1994, **90**, 17-24.

149. P. Suèr and B. Allard, Mercury Transport and Speciation during Electrokinetic Soil Remediation, *Water, Air, and Soil Pollut.*, 2003, **143**, 99-109.

150. Y. B. Acar, R. J. Gale, A. Ugaz, S. Puppala and C. Leonard, *Feasibility of Removing Uranium, Thorium and Radium from Kaolinite by Electrochemical Soil Processing*, Report EK-BR-009-0292 (Final Report-Phase I of EK-EPA Cooperative Agreement CR816828-01-0), Electrokinetics Inc., Baton Rouge, Louisiana, US, 1992.

151. X. Hou, Radioanalysis of ultra-low level radionuclides for environmental tracer studies and decommissioning of nuclear facilities, *J. Radioanal. Nucl. Chem.*, 2019, **322**, 1217-1245.

152. A. Leskinen, S. Salminen-Paatero, C. Gautier, A. Räty, M. Tanhua-Tyrkkö, P. Fichet, T. Kekki, W. Zhang, J. Bubendorff, E. Laporte, G. Lambrot and R. Brennetot, Intercomparison exercise on difficult to measure radionuclides in activated steel: statistical analysis of radioanalytical results and activation calculations, *J. Radioanal. Nucl. Chem.*, 2020, **324**, 1303-1316.

153. P. E. Warwick and I. W. Croudace, Rapid on-site radionuclide screening of aqueous waste streams using dip-stick technologies and liquid scintillation counting, *J. Radioanal. Nucl. Chem.*, 2017, **314**, 761-766.

154. X. Hou, Radiochemical analysis of radionuclides difficult to measure for waste characterization in decommissioning of nuclear facilities, *J. Radioanal. Nucl. Chem.*, 2007, **273**, 43-48.

155. K. Onwubuya, A. Cundy, M. Puschenreiter, J. Kumpiene, B. Bone, J. Greaves, P. Teasdale, M. Mench, P. Tlustos, S. Mikhalovsky, S. Waite, W. Friesl-Hanl, B. Marschner and I. Müller, Developing decision support tools for the selection of “gentle” remediation approaches, *Sci. Total Environ.*, 2009, **407**, 6132-6142.

156. A. Cundy, P. Bardos, M. Puschenreiter, N. Witters, M. Mench, V. Bert, W. Friesl-Hanl, I. Müller, N. Weyens and J. Vangronsveld, Developing Effective Decision Support for the

Application of “Gentle” Remediation Options: The GREENLAND Project, *Remed. J.*, 2015, **25**, 101-114.

157. S. Mobbs, P. Orr and I. Weber, Strategic considerations for the sustainable remediation of nuclear installations, *J. Environ. Radioact.*, 2019, **196**, 153-163.

158. L. Huysegoms and V. Cappuyns, Critical review of decision support tools for sustainability assessment of site remediation options, *J. Environ. Manage.*, 2017, **196**, 278-296.

159. *Strategic Considerations for the Sustainable Remediation of Nuclear Installations*, Report NEA No. 7290, Nuclear Energy Agency (NEA) of the Organisation for Cooperation and Development (OECD), 2016.

160. R. P. Bardos, H. F. Thomas, J. W. N. Smith, N. D. Harries, F. Evans, R. Boyle, T. Howard, R. Lewis, A. O. Thomas and A. Haslam, The Development and Use of Sustainability Criteria in SuRF-UK’s Sustainable Remediation Framework, *Sustainability*, 2018, **10**, 1781.