Lambda Consult

PRELIMINARY ASSESSMENT OF THE APPLICATION OF AN ELECTROKINETIC RING FENCE FOR THE REMOVAL OF RADIONUCLIDES FROM GROUNDWATER AT

FUKUSHIMA DAIICHI NUCLEAR POWER PLANT

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EXECUTIVE SUMMARY

In the summer of 2013, more than two years after the incident with the Fukushima Daiichi nuclear power plant, it was revealed that the plant is leaking radioactive water into the Pacific Ocean. Apart from leakages from storage tanks, rainwater is picking up radioactive contamination on the surface before it flows into the sea. But at this moment the groundwater is the biggest problem at the plant with an estimated daily outflow of 800 tons of water - half of it traveling into the ocean, the other half seeping into the facility's buildings and requiring storage.

The latest plans include an ice wall, a new groundwater pumping system and yet another system to filter radionuclides. But the ice-wall technology is unproven and their costs are estimated at US$ 470 million.

We propose an alternative and less costly technology (about one order of magnitude less) to contain the outflow of radionuclides into the aquifer and the sea by deploying a so-called Electrokinetic Ring Fence (EKRF).

Electrokinetic soil and groundwater remediation methods are based on the transport processes that occur when an electric current is passed through the subsurface. The current is carried by ions, and the anions (negative charged ions) will be transported toward the anode and the cations (positive charged ions such as radionuclides) toward the cathode. This transport of ions in the electric field is electromigration.

Electrokinetic technology can be applied as a passive method in the form of an electrokinetic fence for containment and remediation of groundwater plumes contaminated with e.g. heavy metals and/or radionuclides in ionic form.

Groundwater velocity is one of the major factors in determining whether an EKRF is economically viable from the point of view of electrical power supply. The higher the groundwater velocity the higher the necessary current and voltage between the electrodes and the higher the annual operating cost. The necessary electrical power can be diminished by decreasing the distance between the electrodes: the smaller the distance the less electrical power is needed to deflect and collect the radionuclides in the electrolytes around the electrodes. But smaller distances mean more electrodes and thus higher initial capital costs to build and install the fence. It is therefore necessary to optimize capital cost and annual operating cost.

Electrokinetics is a tried and tested technology for processing soils and groundwater; it has been used for decades as an in-situ method for the remediation of heavy metal contaminated land, mostly at industrial sites. The application as an EKRF,to capture contaminant ions transported by the groundwater has so far not been used for the large scale containment of radioactive groundwater plumes, but several EKRFs have been in operation since 2001 in the Netherlands and in Tokyo since 2005. Recently a proposal has been made for an EKRF in the Netherlands at the front of a 600 m wide groundwater plume contaminated with high concentrations of zinc, which originated from a galvanizing plant. The efficiency of the fence will be ascertained by monitoring radioactivity in the groundwater upstream and downstream of the fence. This can be done by periodic sampling and analysis or by automatic monitoring by telemetric sensors.

The radionuclides are captured in the electrolytes. This very concentrated secondary waste stream has to be removed by Ion Exchange or Electrochemical Ion Exchange systems or another efficient filter system and be disposed off through regular canals.

The reliability of the fence will be assured when a) the equipment is made of high quality material, adapted to a radioactive environment and winter conditions and b) the installation of the electrode casings and electrolyte management system is executed with utmost care under strict control. Control of electrical power system, electrolyte management system and electrodes will be automated.

The EKRF approach that we are proposing for this site is significantly different from the approaches so far envisaged by TEPCO. Based on our experience and in our opinion realistic assumptions, we have prepared a best efforts estimate of the overall cost for this approach together with the annual operating costs:

- Containerized units for electrical power supply, electrolyte management	€ 30 to 40 million
- Electrolyte treatment (removal of radioactive material)	PM
- Cost and installing electrodes and other field appliances	€ 12 to 18 million
- Annual operating cost *	€ 2.5 to 5 million
- Disposal of radioactive material	PM

** This includes the cost of energy which is estimated at 7 to 9 million kWh/year at € 0.12 per kWh. Final cost calculations can be given when all site specific data are known*

The calculations are based on the following assumptions:

- Length of EKRF	1625 m	- Distance between anode-cathode	5 m
- Depth of phreatic groundwater level	2 m	- Number of anode casings	164
- Depth of radioactive contamination	15 m	- Number of cathode casings	163
- Groundwater velocity	20 m / year	- Total current strength	4000-5000 A
- Average mobility of radionuclides	1 m^2 / V year	- Electrical power connection	800-1000 kW/400 V
Soil resistivity	50 Ohmm		
Groundwater resistivity	15 Ohmm		

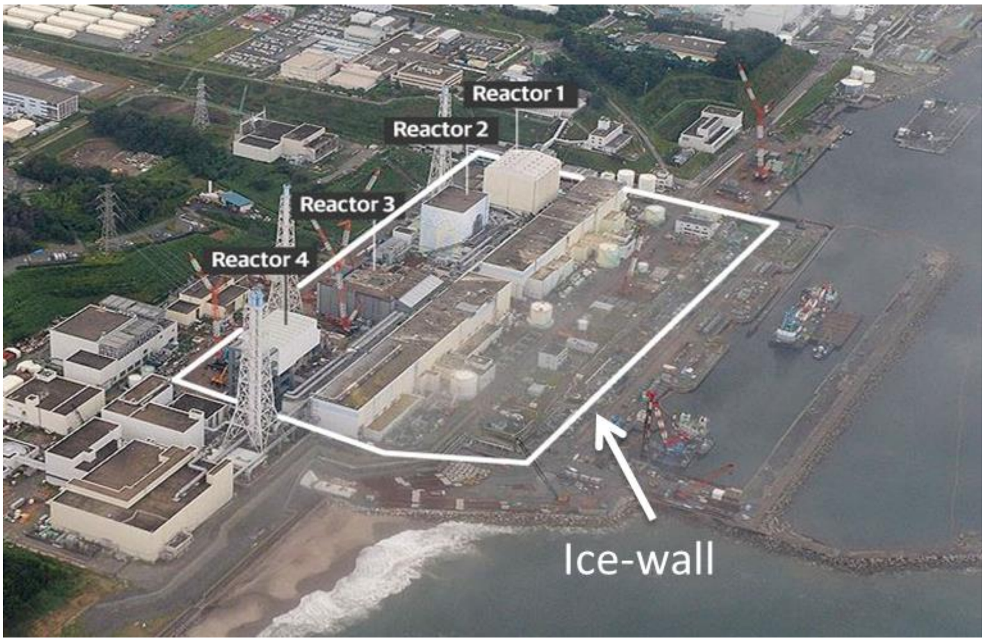
The feasibility of an EKRF at the Daiichi Nuclear Power Plant can be tested beforehand by running a pilot test at the site. Such a test could be accomplished within a period of 6-8 months at an estimated cost of € 250k.

1 INTRODUCTION

On March 11 2011, the Fukushima Daiichi nuclear power plant was hit by a 14-meter tsunami, generated by the Tohoku earthquake. On July 22, 2013, more than two years after the incident, it was revealed that the plant is leaking radioactive water into the Pacific Ocean, something long suspected by local fishermen and independent investigators. In the following months more leaking incidents happened and the current situation has prompted Japanese Prime Minister Shinzō Abe to order the government to step in.

Apart from leakages from storage tanks, rainwater is picking up radioactive contamination on the surface before it flows into the sea. But at this moment the groundwater is the biggest problem at the plant. TEPCO estimates that 800 tons of water flows under the plant daily — half of it traveling into the ocean, the other half seeping into the facility's buildings and requiring storage.

According to insiders the groundwater problem needs to be solved as soon as possible. The latest plans include an ice wall, a new groundwater pumping system and yet another system to filter radionuclides. But the ice-wall technology is unproven and very expensive (see picture).



This picture has been taken form the website of the Guardian Newspaper of 15 October 2013 (http://www.theguardian.com/environment/interactive/2013/oct/15/fukushima-daiichi-nuclear-power-plant-tsunami-cleanup-interactive)

The accompanying text reads: "Revelations that contaminated groundwater is seeping into the Pacific ocean every day prompted the government to announce a 32bn yen underground "ice wall" around reactors 1-4. Groundwater flows down the hills behind the plant and mixes with water used to cool the reactors. Frozen-wall technology is not new, but has never been attempted on such a large scale. Critics say the $470m plan is too costly and impractical".

In view of the problem described above, we propose an alternative and less costly technology to contain the outflow of radionuclides into the aquifer and the sea by deploying a so-called Electrokinetic Ring Fence (EKRF), the principle of which will be outlined in the following pages.

2. Site Characteristics

2.1 General

The coastal Daiichi plant is on an old riverbed; at the land side bordered by a line of forested hills and mountains. Even before the 2011 disaster, rainfall from across the region would funnel toward the plant. Such inflow was rarely a problem, because a piping system collected groundwater and let it flow into the ocean. Minor leaks would sometimes form in buildings built below sea level, but even that water, uncontaminated, was easy to pump out and discharge.

The 14-meter tsunami wave of March 11, 2011, disrupted the plant's groundwater discharge system. Damaged pipes no longer catch the inflow, meaning that the plant lost its first line of defence against water streaming in from the hillsides. Worse, the plant had become a highly contaminated site, and any water that flows under or through the area picks up radioactive contaminants. Groundwater that makes its way into the reactor buildings also mixes with a separate channel of intensely contaminated water that had been used to douse and cool the reactors. The groundwater can no longer simply be discharged into the ocean.

2.2 Hydrogeological setting

Besides the description above, we have no data available on the hydrogeological setting at the site of the plant. Neither do we have any information on groundwater levels and other groundwater related parameters. In view thereof the cost estimate which is given in chapter 4 is based on rough approximations only.

2.3 Contaminant situation

Groundwater at Fukushima power plant has been found to be highly contaminated with radionuclides Strontium-90. Cesium 134, Cesium 137 and Tritium. To date we have no information on the concentrations of these species, only on radiation levels.

3 Electrokinetic Remediation Technology

3.1 Historic background

Examination of the history of electrokinetics in soil will reveal it has its beginnings over 80 years ago. Some highlights are the work carried out by Puri and Anand (1936) who tested the value of applying an electric potential across a soil for removing sodium ions in order to improve the soil quality.

Casagrande was the pioneer in stabilizing clays by electroosmosis and his attempts to remove water from lime sludge in the 1930s and 1940s. In the 1950s Collopy patented the use of electromigration for reclaiming saline soils. The beginning of the 1980s marked the interest both in Europe and in the USA of using the technology for the removal of toxic ions from the soil. Most of the early work and in particular the field experiments, however, were inconclusive due to the failure to manage the electrochemical changes in the soil around the electrodes. The breakthrough came in 1987 with Lageman, Pool and Seffinga from the Dutch company Geokinetics. They focused on electromigration and patented the use of circulating electrolytes around the electrodes and the use of ion permeable wells to manage and control the anolyte and catholyte. Since then Lageman c.s. have executed dozens of electrokinetic in-situ remediation projects in the Netherlands, Italy and the USA, recovering contaminants such as lead, copper, mercury, zinc, arsenic, cadmium, chromium etc.

Experiments with electrokinetic migration and removal of radioactivity from soil was carried out in the early 1990s by Pamuckcu et al., followed by Maes et al. at the turning of the millennium. The most recent studies on electrokinetic removal of radionuclides from the soil during the 2000s have been carried out by Kim et al. and Korolev et al., the latter mostly as a result of the accident at the Chernobyl Nuclear Power Plant in 1986.

The number of papers published in the late 1980s in this field was less than 20, but during the 1990s more than 400 papers have been published. During the 2000s this number has more than doubled. In the meantime a worldwide network has been formed. At present this network constitutes about 100 teams mainly from Europe and Northern America, but also teams from Asia, Southern America and Australia have joined the network.

3.2 Principle of electrokinetic remediation

Electrokinetic soil and groundwater remediation methods are based on the transport processes that occur when an electric current is passed through the subsurface. The transport mechanisms of major importance to the electrochemical soil remediation methods are electromigration and electro osmosis. In the soil the current is carried by ions, and the anions (negative charged ions) will be transported toward the anode and the cations (positive charged ions) toward the cathode. This transport of ions in the electric field is electromigration. Electro osmosis is movement of pore water in the soil in the applied electric field.

The methods where an electric field is applied to a soil to remove chemical species are variably called e.g. electrokinetic remediation, electro-reclamation, electrokinetic extraction, electrokinetic soil processing, electrochemical decontamination or electrodialytic remediation.

3.3 Applications of Electrokinetic Technology

3.3.1 In situ site remediation

Electrokinetic technology can be applied as an active in situ method to clean-up sites where land is contaminated with e.g. heavy metals and/or polar contaminants and radionuclides.

The key elements are as follows:

- ion-permeable electrolyte casings are placed in the contaminated media and connected to a electrolyte management system (f*ig. 1*). Each casing has an electrode inside. Together, these form alternating rows of anodes and cathodes. Electrolyte is circulated in a closed loop between the electrode casings and the management system;
- the electrodes are then energized. Electrolysis of water in the electrolyte results in the formation of H^+ ions at the anodes and OH^- at the cathodes. These ions are then made to migrate through the casing into soil to generate a temporary and localized pH shift which desorbs contaminating ions. Acids are not pumped into the soil;
- once desorbed, the contaminating ions migrate under the influence of the applied potential (electro migration) to the electrodes. Negatively charged anions go to the positive anodes, positively charged cations go to the negative cathodes). Here they pass through the electrode casing walls and are taken up by the circulating electrolytes;
- careful management of the pH and other electrolyte conditions within the electrode casings is the critical element in controlling system performance;
- in general contamination is removed from the electrolytes by precipitation and filtration. Contamination can also be selectively recovered from the circulating electrolytes by Ion-Exchange (IX) and/or Electrochemical Ion-Exchange (EIX) or deploying an EnviroCell.

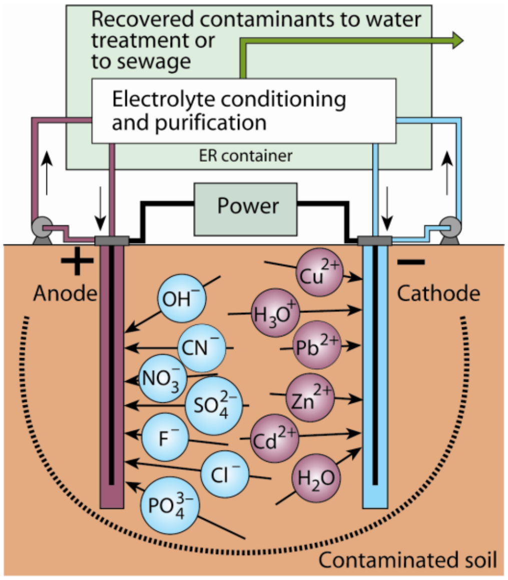


Figure 1.Schematic setup of electrokinetic system for in-situ remediation

In the present situation in-situ electro-remediation of the contaminated soil is no option. There are two main reasons:

- Electrokinetic laboratory tests with Cs showed that Cs is very strongly adsorbed to clay minerals and can only be mobilized against high energy input.
- Above ground buildings and below ground infrastructure and foundations hamper the installation of a dense electrode network.

3.3.2. In-situ groundwater plume containment by an Electrokinetic Ring Fence

However, the technology can be deployed in the form of an electrokinetic fence as a passive in-situ clean-up method for containment and remediation of groundwater plumes contaminated with e.g. heavy metals and/or radionuclides in ionic form. The basic setup consists of a row of alternating anode and cathode electrodes bordering a high-concentration area or polluted groundwater plume. The row of vertical electrodes is preferably set perpendicular to the prevailing groundwater flow direction, while the depth of the electrodes coincides with the lowest depth where pollutants are found. The heavy metal or radionuclide ions transported with the groundwater are captured in the electrolytes and subsequently removed from the electrolytes above ground (*fig. 2*). There is no disturbance of the groundwater flow regime.

The efficacy of an electrokinetic fence can in general be defined as:

$$N_d = \frac{\text{number of charged particles captured by the fence}}{\text{number of charged particles entering the fence zone}} \quad \text{or} \quad N_d = \frac{C_b - C_e}{C_b} * 100,$$

wherein:

N_d = efficacy (%)

C_b = concentration of ions in front of the fence (µg/l)

C_e = concentration of ions after the fence (µg/l)

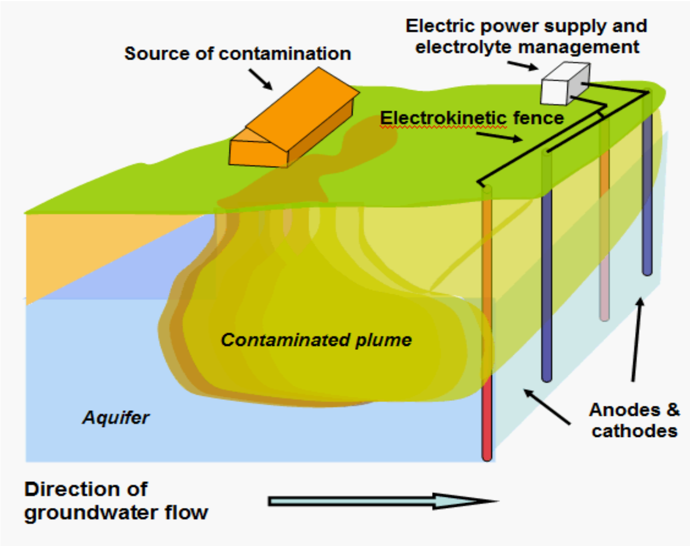


Figure 2. Schematic representation of electrokinetic fence for in-situ containment

Electrodes can also be installed in a ring around a storage site with contaminated and/or radioactive material.

The optimal configuration of an Electrokinetic Ring Fence (EKRF) can be determined by computer simulations that calculate the velocity of the charged particles coming toward the fence for each value of the relevant physical/chemical and geohydrological parameters. Important parameters in this respect are:

Symbol	Parameter	Dimension
V_{gw}	Groundwater velocity	m/year
K_{ek}	Electrokinetic mobility	m^2/Vs
Φ	Electric potential	V
K	Specific electric conductivity of the soil	S/m
L	Length of the electrodes	m
D	Distance between the electrodes	m

It will not be difficult to realize that the groundwater velocity is one of the major factors in determining whether an EKRF is economically viable or even possible from the point of view of electrical power supply. The higher the groundwater velocity the higher the necessary voltage between the electrodes and the higher the necessary current per meter electrode and clearly the higher the annual operating cost. There is however, a limit to the current which can be supplied to the electrodes. The method to diminish the necessary electrical power is to decrease the distance between the electrodes: the smaller the distance the less electrical potential is needed to deflect and collect the ions in the electrolytes and the less electrical current has to be supplied by the electrodes. But smaller distances mean more electrodes and thus higher initial capital costs to build and install the fence. It is therefore necessary to optimize capital cost and annual operating cost.

Optimization is accomplished by first using a simple analytical formula to calculate preliminary values of the necessary voltage and current as a function of the distance between the electrodes, the soil and water resistivity, ion mobility and groundwater velocity. These values are then validated in a computer program, which calculates the flow path of the ion when entering the zone of the fence (*fig.3*). On the basis of these results one can run several simulations to obtain the optimum values for installation and operating costs of the electrokinetic fence.

Figure 3 depicts the results of a simulation with assumed data as given in section 4.1. It can be observed that at a groundwater velocity of 20 m/year about 25% of the Strontium ions entering the fence area near the anode are captured at an applied potential of 38 V between the electrodes. At 75 V about 50% of the Strontium ions are captured and at 150 V, 100% of the Strontium ions are captured. The distance between the electrodes is 5 m and the current strength is a function of the required voltage over the electrodes and the resistivity of the soil.

Similar simulations can be made for Cs and other radioactive species. The actual potential of the fence will be set on the basis of the least mobile ion.

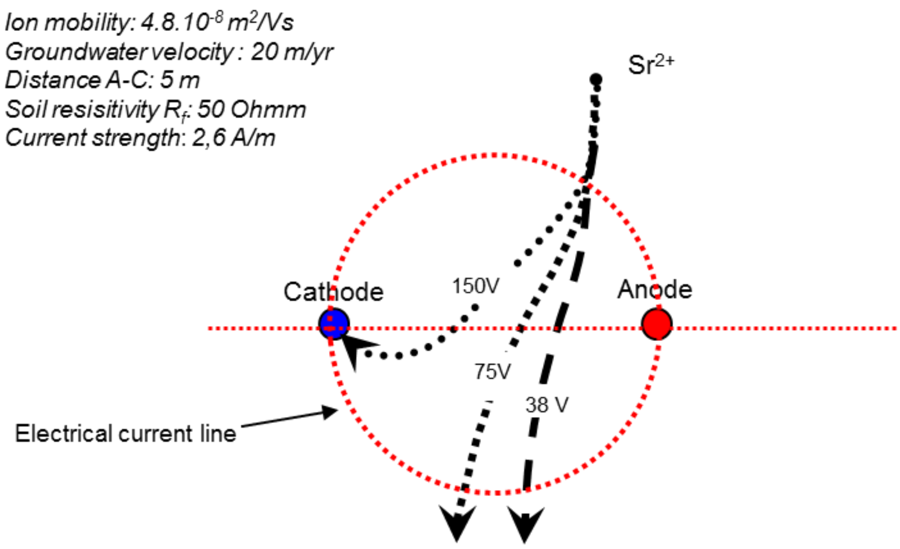
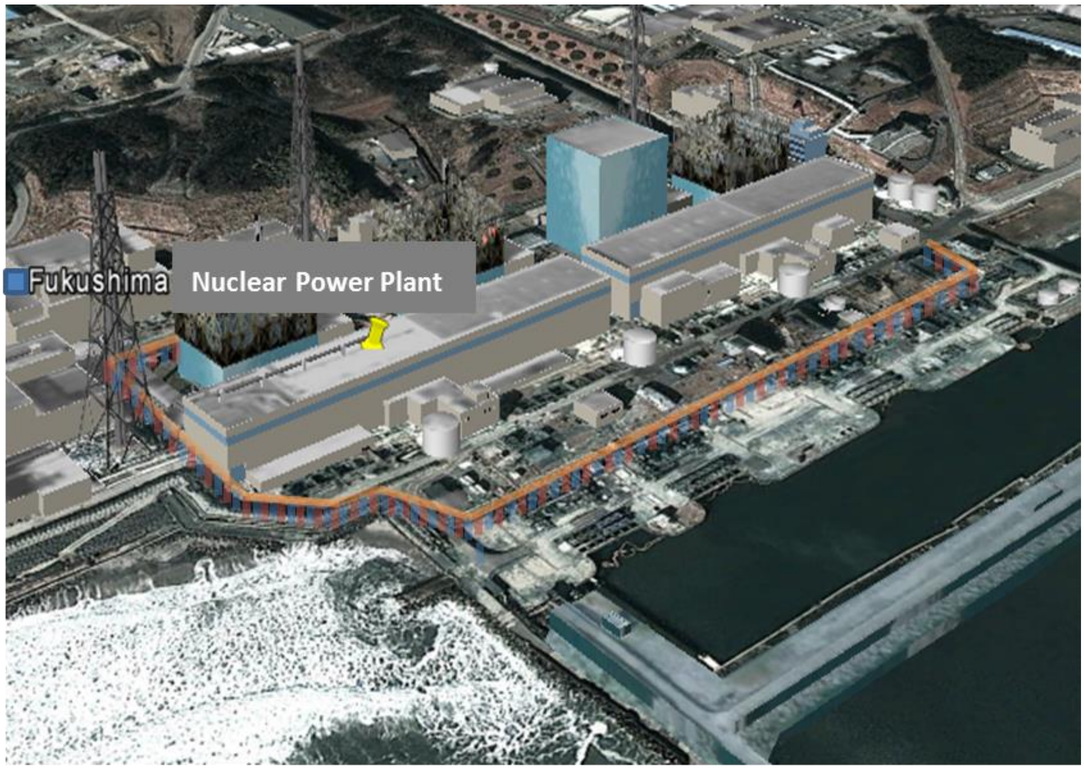


Figure 3. Computer simulation of Strontium ion entering the fence zone (top view)

4. Feasibility of an EKRF at the site of the power plant

4.1 Field Set-up

We have used the geometry of the “ice wall” as a basis for preliminary calculations of capital cost and annual operational cost of an EKRF (see picture below).



Location of Electrokinetic Ring Fence

Vertical Anode electrode

Vertical Cathode electrode

The calculations are based on the following assumptions:

- Length of EKRF	1625 m	- Distance between anode-cathode	5 m
- Depth of phreatic groundwater level	2 m	- Number of anode casings	164
- Depth of radioactive contamination	15 m	- Number of cathode casings	163
- Groundwater velocity	20 m / year	- Total current strength	4000-5000 A
- Average mobility of radionuclides	1 m^2 / V year	- Electrical power connection	800-1000 kW/400 V
- Soil resistivity	50 Ohmm		
- Groundwater resistivity	15 Ohmm		

The final design and electrode configuration together with the electrical power requirements can be given when data on the soil and groundwater parameters listed above are known.

4.2 Performance

Electrokinetics is a tried and tested technology for processing soils; it has been used for decades as an in-situ method for the remediation of heavy metal contaminated land, mostly at industrial sites. It has the advantage of being effective in low permeability strata for which other in situ techniques are less effective.

But in this case as explained in the foregoing chapter, it should be effectively applied as an EKRF, capturing the contaminant ions transported by the groundwater. This approach has so far not been used for the large scale containment of radioactive groundwater plumes, but an EKRF has been in operation in Tokyo since 2005 at the site of an industrial area to prevent chromium migrating to the adjacent downstream site of a neighbouring company. Recently a proposal has been made for an EKRF in the Netherlands at the front of a 600 m wide groundwater plume contaminated with high concentrations of zinc, which originated from a galvanizing plant..

Another EKRF has been installed and tested in the Netherlands during the period 2001-2007 in the form of an electrokinetic biofence. This fence has been installed near the building of a chemical laundry to disperse electrokinetically dissolved polar nutrients in the groundwater in order to enhance reductive dechlorinization of present perchloroethene (PCE), trichloroethene (TCE), Cis 1,2-dichloroethene (C-DCE) and vinylchloride (VC). The fence acted as a temporary safety measure to avoid further migration of the contamination from the source area underneath the building to the plume area.

Electrical power requirements can be calculated beforehand and adjusted during operation by automatic measurement of groundwater levels before and after the fence in monitoring wells. The change in hydraulic gradient and hence groundwater velocity can be used to increase or decrease the necessary electrical power supply.

The efficiency of the fence will be ascertained by monitoring radioactivity in the groundwater upstream and downstream of the fence. This can be done by periodic sampling and analysis or by automatic monitoring by telemetric sensors

Another important item is the generation of a secondary waste stream of radionuclides in the electrolytes. They have to be removed by IX or EIX systems or another efficient filter system and be disposed off through regular canals.

Finally, the total period required to keep the fence operational depends on the contaminant situation at the source area, i.c. the nuclear power plant. As long as the groundwater upstream of the fence contains too high levels of Becquerel’s per liter, the fence has to stay activated.

4.3 Reliability and maintenance

The reliability of the fence will be assured when a) the equipment is made of high quality material, adapted to a radioactive environment and winter conditions and b) the installation of the electrode casings and electrolyte management system is executed with utmost care under strict control. Control of electrical power system, electrolyte management system and electrodes will be automated. Maintenance of the system is rather easy as all components are easily accessible

4.4 Availability

The electrokinetic system consisting of shielded containerized electrical power, electrolyte management and treatment units, as well as electrode casings cannot be procured 'off the shelf’ but have to be designed and built. Manuals and drawings are available and need adaptation to a radioactive environment only. There are in Japan several companies that have practical experience with electrokinetic projects in the past and under proper supervision the electrokinetic installation outlined in section 4.1. could be built in 6-8 months. During this period borings for the electrode casings can already be executed with a small easy to handle drilling machine.

4.5 Risk to workers and public safety

The site is part of a nuclear disaster area with very strict safety procedures. Non-Tepco personnel at the site will have to be trained to work in a high risk environment.

4.6 Pilot Project

The feasibility of an EKRF at the Daiichi Nuclear Power Plant will have to be tested beforehand by running a pilot test at the site. The length of the fence can be confined to 10-15 m. Assuming a groundwater velocity of around 20 m/year implicates that radionuclide-ions will travel at a velocity of approximately 1.5 m/month. After 6 months a distance of around 9 m would have been covered, which would be enough to compare radioactive radiation and/or concentration upstream and downstream of the fence.

Such a test could be accomplished within a period of 6-8 months at an estimated cost of € 250k.

5 PRELIMINARY COST ESTIMATE

The EKRF approach that we are proposing for this site is significantly different from the approaches so far envisaged by TEPCO. Based on our experience and in our opinion realistic assumptions, we have prepared a best efforts estimate of the overall cost for this approach together with the annual operating costs:

- Containerized units for electrical power supply, electrolyte management	€ 30 to 40 million
- Electrolyte treatment (removal of radioactive material)	PM
- Cost and installing electrodes and other field appliances	€ 12 to 18 million
- Annual operating cost*	€ 2.5 to 5 million
- Disposal of radioactive material	PM

** This includes the cost of energy which is estimated at 7 to 9 million kWh/year at € 0.12 per kWh*

Final cost calculations can only be given when all site specific data are known.

For Lambda Consult,
Reinout Lageman, January 2014