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Article

Design Scheme to Develop Integrated Remediation Technology: Case Study of Integration of Soil Flushing and Pneumatic Fracturing for Metal Contaminated Soil

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In remediation of the contaminated soil, it requires to select at least more than two remediation technologies depending on the fate and transport phenomena through complicated reactions in soil matrix. Therefore, methodologies related to develop the integrated remediation technology were reviewed for agricultural soils contaminated with heavy metals. Pneumatic fracturing is necessary to implement deficiency because soil washing is not effective to remove heavy metals in the subsurface soil. But it needs to evaluate the characteristics such as essential data and factors of designated technology in order to effectively apply them in the site. In the remediation site, the important soil physical and chemical factors to be considered are hydrology, porosity, soil texture and structure, types and concentrations of the contaminants, and fate and its transport properties. However, the integrated technology can be restrictive by advective flux in the area which remediation is highly effective although both soil washing and pneumatic fracturing were applied simultaneously in the site. Therefore, we need to understand flow pathways of the target contaminants in the subsurface soils, that includes kinetic desorption and flux, predictive simulation modeling, and complicated reaction in heterogenous soil.

Key words : Integrated remediation, Soil washing, Pneumatic fracturing, Heavy metals

Introduction

Contamination of soils with heavy metals is widespread and poses a long-term risk to ground water quality and ecosystem health. The development of desirable technologies for treating contaminants is a critical step in the effort to cleanup the hazardous waste sites because the chemical form of the contaminant can influences its solubility, mobility, and toxicity in soil (McBride, 1994).

Remediation technologies are classified as three types as follows (EPA, 1995): soil can be excavated from the ground and be either treated or disposed (ex-situ); soil can be left in the ground and treated in place (in-situ); or soil can be left in the ground and contained to prevent the contamination from becoming more widespread and reaching plants, animals, or humans (Containment). Three primary strategies used separately or in conjunction to remediate most sites are (EPA, 1998): destruction or alteration of contaminants, extraction or separation of contaminants from environmental media, and immobilization of contaminants (Fig. 1).

However, selection of treatment methods should use the most effective contaminant transport mechanisms to arrive at the most effective treatment scheme. Regardless of the strategy employed, most in situ systems designed for the reduction or removal of contaminants incorporate the introduction of some treatment media. Physical /chemical treatment uses the physical properties of the contaminants or the contaminated medium to destroy (i.e., chemically convert), separate, or contain the contamination. Therefore, more than one treatment technology should be combined at a single site to reach the desirable cleanup level enforced by regulation or land use because solitary technologies may only treat one phase of the contamination when, in fact, the contamination is often spread through multiple phases and zones (EPA, 1998).

Many scientific studies have generated a large body of scientific information on fate of heavy metal governed by fundamental chemical reactions between metal constituents and soil and residual components. Heavy

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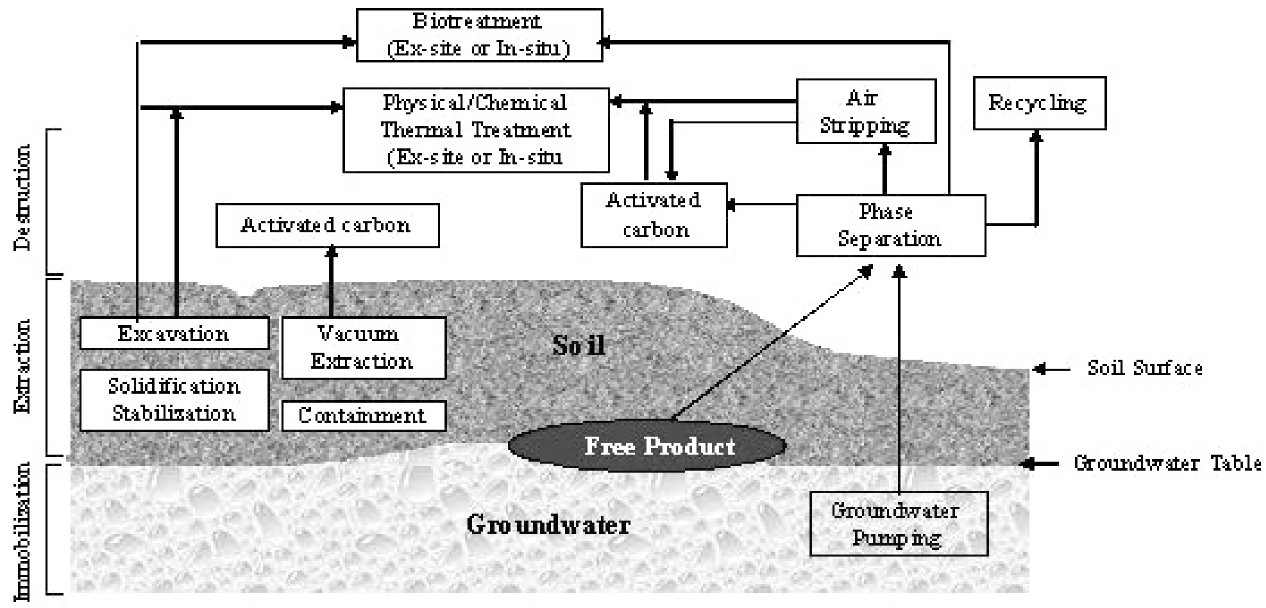


Fig. 1. Classification of remedial technologies by function based on the general three approaches.

metals which are sorbed to soil organic matter regulates partitioning of heavy metals between solution and solid phases in soils. Sorption and many metal precipitation processes are highly pH dependent with increased sorption with pH. And heavy metals can form sparingly soluble phosphate, carbonates, sulfides, and hydroxides. And so it must evaluate the fate of contaminants including adsorption and desorption behavior, mass flow of heavy metals with respect to spatial soil physical property, criteria of design method prior to the development of strategy for integrated remediation technology (Allen et al., 1991).

The primary objectives of this review were to provide information about the intrinsic design scheme and relevant factors needed for developing the integrated remediation scheme which was focused on incorporation of pneumatic fracturing (hereafter PF) into in situ soil flushing (hereafter SF) for heavy metal contaminated soil, as of the effective and cost-saving physical-chemical remediation technologies.

Status of the Heavy Metals in Soil

Soils can be a large sink for anthropogenic and natural heavy metals. Soil contamination is either solid or liquid phase mixed with the naturally occurring soil (Andreas et al., 2003). Important metal types that are released by mining and smelting operations are sulfides and oxides. Their dissolution and phase transformation determines the initial fate of these heavy metals in the soil.

Usually, heavy metals in the soil are physically or chemically attached to soil particles, or, if they are not attached, are trapped in the small spaces between soil particles. The chemical behavior of heavy metals in soils is controlled by a number of processes, including metal cation release from contamination source materials (e.g., fertilizer, sludge, smelter dust, ammunition, slag), cation exchange and specific adsorption onto surfaces of minerals and soil organic matter, and precipitation of secondary minerals (Manceau et al., 2000; McBride, 1999; Morin et al., 1999). Also, the physical properties of the heavy metal contaminated soils hinder the application of the technologies because of high bulk density and low hydraulic conductivity resulting in difficulties in delivering the treatment fluid.

Statement of problems in application of remediation technologies

In remediation of heavy metals in soils, it requires soil flushing to extract and recover soil solution from soils. But SF is not effective in subsurface soils because of soil physical properties such as high bulk density and low permeability which that result in inconsistent removal rates although treating fluid is desirable. That is a significant technical challenge how to overcome these limitations in the field to effectively achieve the remediation goal by in situ remediation efforts. The remediation design should consider the difficulties and limitations in selecting the technology to enhance permeability in soils prior to application of displacing reagents using soil flushing system. Therefore, it needs supplementary or pertinent technology to improve soil physical properties which can control the effectiveness of in situ treatment technologies. This can be interpreted as integration of remediation technology that can include more than two technologies for cleanup the contaminated soils. Table 1 shows the typical in situ remediation technologies for heavy metal contaminated soils (EPA, 2005).

Selection of technologies should use the most effective contaminant transport mechanisms to arrive at the most effective treatment scheme. Especially for heavy metals strongly adsorbed onto soil particle surface, extraction and removal technologies can, however, encounter difficulty in delivery of treatment reagents to displace and remove the contaminants due to poor accessibility to the contaminants regardless of treating reagents in the field. Also low permeability or heterogeneous soils hinders the mobility of heavy metals by reactions of flushing fluids with soil along with the unwanted spread of contaminants and closed fracture in non-clayey soils during fracturing.

The Solution

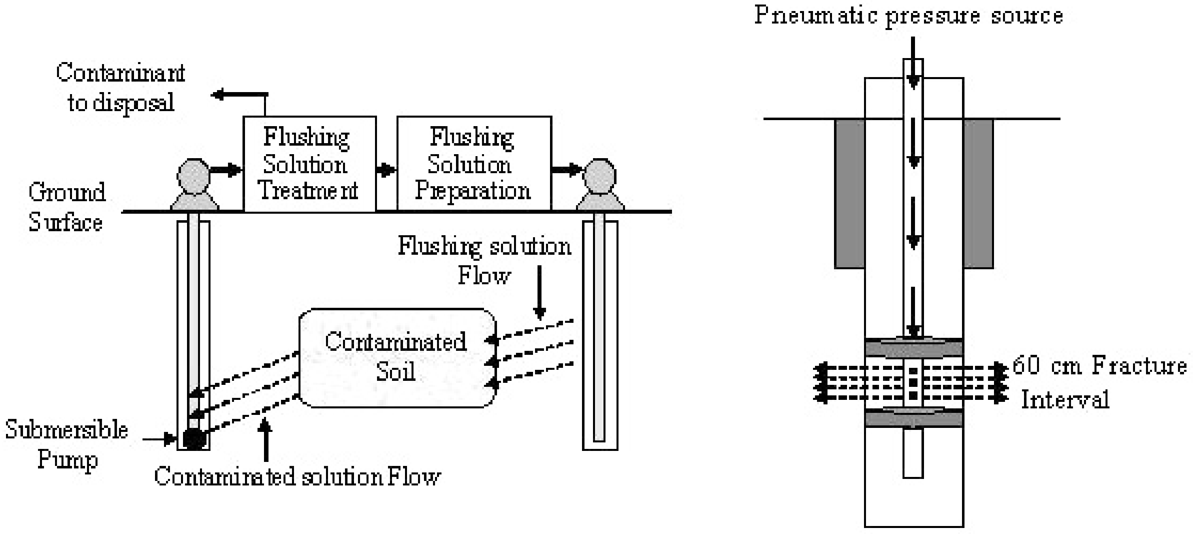
The main advantage of in situ treatment is that it allows soil to be treated without being excavated and transported, resulting in potentially significant cost savings. Physical/chemical treatment is typically cost effective and can be completed in short time periods (in comparison with biological treatment). Generally, no single technology can remediate an entire site. Several treatment technologies are usually combined at a single site to form what is known as a treatment train. SVE can be integrated with ground water pumping and air stripping to simultaneously remove contaminants from soil.

Physical/chemical treatment is typically cost effective and can be completed in short time periods in comparison with biological treatment. Equipment is readily available and is not engineering or energy-intensive. Soil flushing is the extraction of contaminants from the soil with water or other suitable aqueous solutions while pneumatic fracturing creating cracks beneath the surface in low permeability and over-consolidated sediments can open new passage ways in difficult soil conditions that increase the effectiveness of many in situ processes and enhance extraction efficiencies (Fig. 2). However, there has been very little commercial success with soil flushing because of delivery problems in low permeable soils regardless of environmentally compatible extraction fluids to remove heavy metals from the soil particle surfaces in the filed.

Integration of remediation technology should minimize the cost of achieving risk-based endpoints by selecting treatment trains or technology combinations that, when coupled together, work in a synergistic manner. For example. Dupont developed the integrated in situ remediation technology for many contaminated sites which had poor accessibility to the contaminants and

Table 1. Typical in situ remediation technologies for heavy metal contaminated soil.

Technology	Media	Types of contaminants	Description	Treatment Tech.
Electrokinetic Remediation of Heavy Metals and Radionuclides	Soil	Heavy metals	Electrical current is supplied between two electrodes, ions of contaminant will be attracted to one of the electrodes	Electrokinetic Separation
Encapsulation of Hazardous Wastes	Liquid, slurry, solid waste	Metals, inorganics	Encapsulation of wastes	Solidification /Stabilization
In Situ Vitrification of Contaminated Soils	Soil	Heavy metals	Immobilization	Solidification /Stabilization
Mitigation Barrier Covers	Arid soils	Soluble metals	Containment/ Treatment	Passive/Reactive Treatment Walls
Polyethylene Encapsulation of Radionuclides and Heavy Metals	Aqueous salt and concentrate, salt cake, sludge, ash, ion exchange resin in tanks	Toxic metals (e.g., Cr, Pb, Cd)	Encapsulation	Solidification /Stabilization
Remediation of Metals Contaminated Soils by Ligand- Based Extraction Technology	Soil	Pb, Hg, Cr	Density classification followed by extraction to remove metals	Separation, Chemical Extraction



(A) In situ Soil Flushing

(B) Pneumatic Fracturing

Fig. 2. System diagrams of SF(A) and PF(B).

difficulty in delivery of treatment reagents have rendered existing in situ treatments such as bioremediation, vapor extraction, and pump and treat rather ineffective when applied to trichloroethene(TCE)-contaminated soil.

The integrated in situ technology established geotechnical methods to install degradation zones directly in the filed, and electro-osmosis was utilized to move the contaminants back and forth through those zones until the treatment is completed. Thus, the integrated technology should treat heavy metals with expectation of benefits over existing ones in many aspects including environmental impacts, cost effectiveness, treatment flexibility, and breadth of applications.

Potentially applicable remediation technologies for heavy metal contamination sites are presented in Table 2 (EPA, 2004). These available in situ technologies or treatment approaches for heavy metal contamination use chemical reduction and fixation for remediation (e.g., geochemical fixation, permeable reactive barriers (PRBs), and reactive zones). Other types of in situ approaches under development include enhanced extraction, electrokinetics, biological processes that can be used within PRBs and reactive zones, phytoremediation, and natural attenuation.

Treatment technologies commonly used for extraction and separation of heavy metals from soil treatment are thermal desorption, soil washing, and solvent extraction, or some combination of these technologies. In low permeability soils or heterogeneous soils contaminated with heavy metals the approach involves the synergistic coupling of PF with in situ SF in the treatment zones that are installed directly in the contaminated soils to form an integrated in-situ remedial process.

In EPA's site demonstration program in 1992, PF tested with hot gas injection and extraction indicated that PF increased the effective vacuum radius of influence nearly threefold and increased the rate of mass removal up to 25

Table 2. Typical technologies of its characteristics for treatment of heavy metals.

Type	Technology	Use Rating	Applicability	Reliability	Technology Function
in-situ	Electrokinetic Separation	Limited	Average	Average	Extract
	Soil Flushing	Limited	Better	Average	Extract
	Solidication/ Stabilization	Limited	Better	Average	Immob.
ex-situ	Chemical Extraction	Limited	Average	Average	Extract/ Destruct
	Chemical Reduction /Oxidation	Limited	Average	Better	Extract
	Separation	Limited	Average	Average	Extract
	Soil Washing	Limited	Average	Average	Extract
	Solidification/ Stabilization	Limited	Average	Average	Immob.

times over the rates measured using conventional extraction technologies (DOE, 1995). And a pilot demonstration of pneumatic fracturing was sponsored by DOE at Tinker AFB in 1993 pneumatic fracturing increased the average monthly removal rate by 15 times. Therefore, the general concept is to use SF to displace and transport heavy metals from the soil into treatment zones where the heavy metals are removed from the pore water and soil particle surfaces by exchange and miscible displacement after fracturing is completed by PF. Briefly, it consists of the following components:

a) create permeable fractures in close proximity sectioned through the contaminated soil region, and turn them into desorption/migration zones by introducing appropriate flushing fluid.

b) find and utilize appropriate flushing fluid to displace and transport heavy metals from the soil into the treatment zones to minimize complications associated with long-term operation of unidirectional soil flushing processes.

Conceptual design procedures

In optimized designing the integrated remediation technology a remedial strategy and system design can be formulated by a through understanding of the subsurface environment for system design. Design of the remediation technology is conducted in parallel with the other continuing design process rather than in series about the various cleanup technologies, the design requirements, and procurement and planning needs. The tasks involved in this procedures are project planning and support, data acquisition, analytical support and data validation, data usability evaluation, treatability study and pilot testing, preliminary design, intermediate design, prefinal design, and post remedial design support. Generally to develop the new remediation technology, it requires the following procedures as shown in Fig. 3.

Upon completion of the preliminary screening and geotechnical testing, pilot testing is typically conducted for further performance evaluation and to provide a design basis for a full-scale system (Gale, 1982). The pilot test plan of the integrated remediation of SF and PF should incorporate the following steps:

Area selection - It is generally preferred to test within the contaminated zone to reduce the impact of lateral heterogeneities and to collect data on contaminant recovery rates prior to and after fracturing.

Fracture point installation - The fracture intervals and locations are selected to coincide with the target zone of contamination within a layered setting.

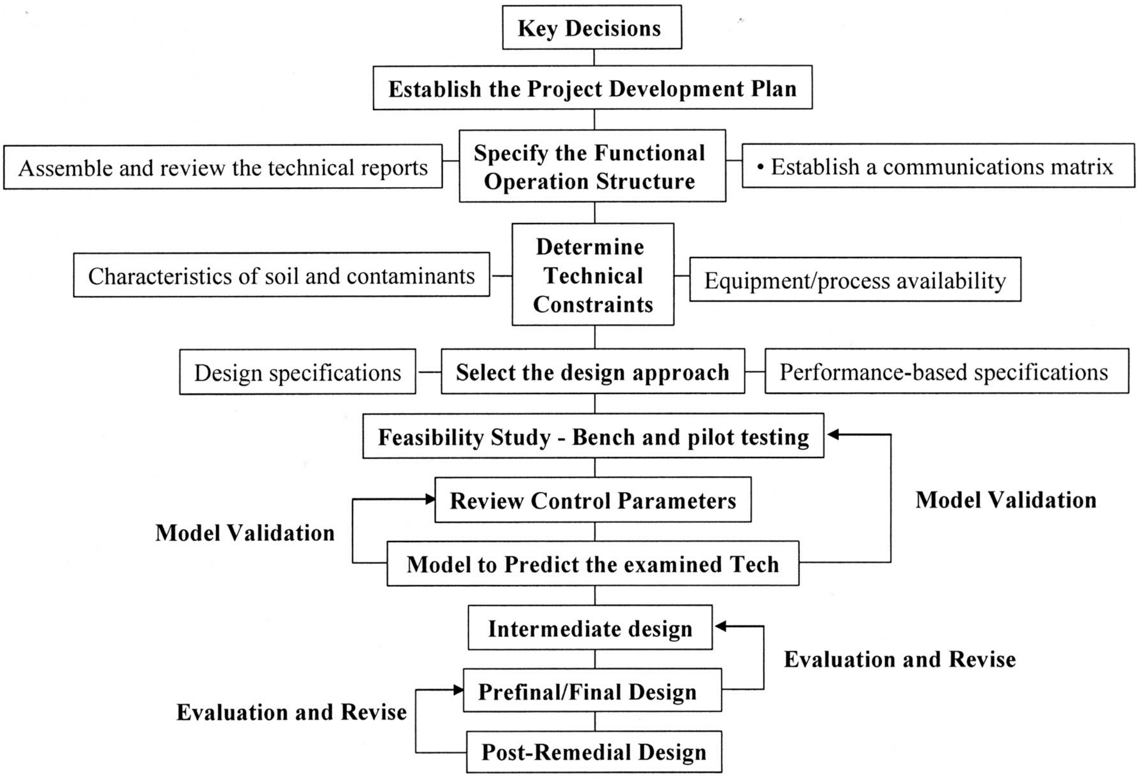


Fig. 3. Generic schedules to develop the remedial technology for a contaminated site.

Test method and monitoring - Pilot testing requires fracture aperture and spacing, fracture orientation, and enhancement of fluid movement.

Baseline permeability/mass recovery estimation - The two general approaches for analyzing flow in fractured media include the equivalent porous medium and the dual porosity approaches.

Compatibility of the flushing solution - Geochemical reactions alter contaminants concentration and subsurface contaminant transport during soil flushing.

With these preliminary screening and pilot testing, design of a full-scale fracturing system can be initiated.

Technical evaluation

Fig. 4 shows the in situ soil flushing and pneumatic fracturing which are considered to be adopted for design of proposed integrated remediation technologies for heavy metal contaminated soils. SF-PF integrated remediation technology will consist of the processes such as displacement of the target element by applying displacing agent and removal of displaced solution filled in soil pore channel induced by PF which can provide the highest possible flow rate. The success of the SF operation will depend on the delivery and movement of the flushing solution through the contaminated zone, and the complete recovery of the elutriate.

The flushing solutions used for SF may consist of one or more of the following: plain water, surfactants and/or cosolvents, acids or bases, reductants/oxidants, chelants, or solvents. The SF process accelerates one or more geochemical reactions that alter contaminants concentration and subsurface contaminant transport mechanisms are accelerate during soil flushing.

The fracture can change the flow paths and patterns of pressure, flux, and travel time in the subsurface. Flux and travel time, however, are more important than pressure for environmental applications. Travel times is significant because some estimates of remediation are based on the number of pore volumes, and the travel time is a measure of the time required to exchange one pore volume within that contour. Fracture size is an important design consideration (Schuring, 1994)

PF applicable for removal of chemicals in low permeable formations is an enhanced technique that physically alters the contaminated soil physical properties to increase the efficiency of soil contaminant extraction and other technologies in low permeability and over-consolidated soil conditions that would otherwise be difficult to treat.

Successful fracturing of a geologic formation with a gas requires that two basic operational conditions be met (Hubbert and Willis, 1957; King, 1993). First, the gas that can not be dissolve in the soil subsurface must be injected at a flow rate that exceeds the ability of the formation to receive the air, i.e., the flow rate must be greater than the native permeability of the formation. Second, the gas must be injected at a pressure that equals or exceeds the in situ geostatic stresses at the depth of injection. Normal PF operation can make 15 to 25 fractures per day with a fracture radius of 4 to 6 meters to a depth of 15 to 30 meters. For longer remediation programs, refracturing efforts may be required at 6- to 12-month intervals.

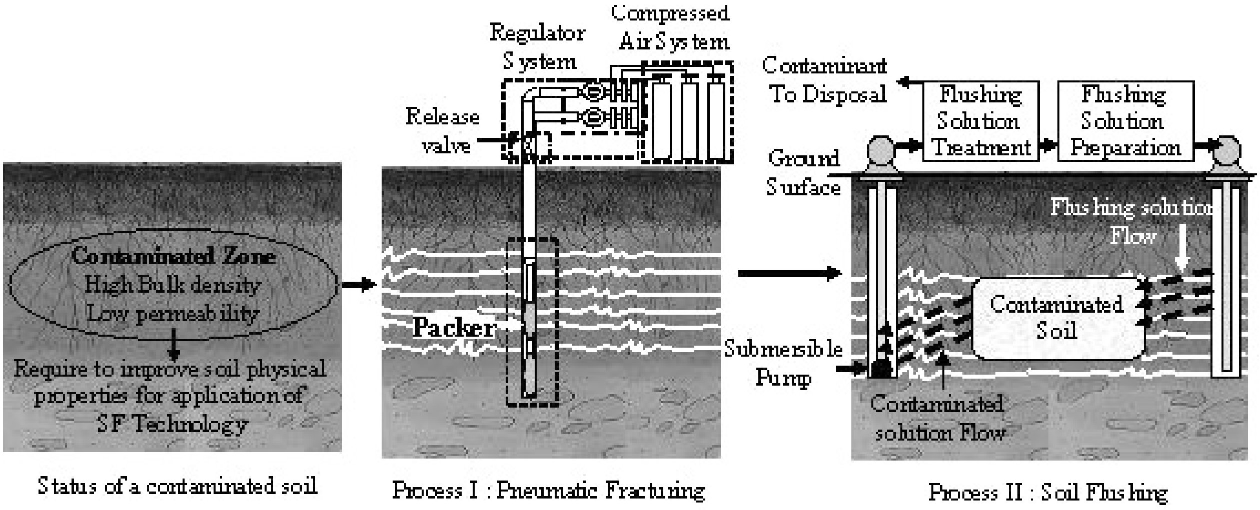


Fig. 4. System configurations of SF and PF and proposed procedures of the integrated SF and PF technology for heavy metal contaminated soils.

Consideration and minimum critical success factors

Solute transport varies depending on macropore continuity and tortuosity developed by fracturing. Given the dynamic nature of mass transport, a given quantity of mass of liquid and dissolved contaminants may pass the surface area of the fractures, in a unit time interval. However, diffusion-controlled mass transfer will influence the time required to reach the cleanup standards. The displacement of heavy metals through soil is strongly affected by adsorption processes (van Genuchten, 1976; Sparks et al., 1980).

Important soil physical and hydrological factors such as permeability, porosity, soil texture and structure, and contaminant type and concentration, and their anticipated fate and transport, are fully evaluated with respect to the design of the desirable and specific technology (AATDF, 1997; Testa, 1994; Testa and Winegardner, 2000). Also following factors should be critically reviewed to optimize the design of the integrated remediation technology,

Clay content and/or particle size distribution : Clay and particle size distribution affect air and fluid flow through contaminated media.

Amount and intervals of compressed air : permeability is a measure of the ease of air flow through soil and is a calculated value.

Hydraulic conductivity/water permeability : Hydraulic conductivity is a measure of the ease of water flow through soil, typically calculated as a function of permeability or transmissivity.

Washing/flushing solution components and dosage of additives : The type and concentrations of additives for a treatment application are determined based on site and contamination characterization, treatability and performance tests, and operator experience.

These site related critical success factors for SF can be summarized as shown in Table 3.

In designing fracturing systems, it is unclear how much and easily the fractures can move the contaminant through soils because it may be very difficult to define a distance of influence for air propagation due the air channel distribution and the variability in the cohesiveness of subsurface soils (Suthersan, 1999). The amount of pressure required to initiate pneumatic fractures is dependent on the cohesive or tensile strength of the formation, as well as the overburden pressure (dependent upon the depth and density of the formation (Bohler et al., 1990, Lundegard, and Andersen, 1993). The pressure required to initiate a fracture in a borehole depends on several factors, including confining stress, toughness of the enveloping formation, initial rate of injection, size of incipient fractures, and pores or defects in the borehole wall.

Critical factors related to fracturing technique include

Table 3. Site related critical success factors for SF.

Critical Success Factor Site Related	Basis	Data Needs
Dominant Contaminant Phase and Equilibrium Partitioning Coefficient	Contaminant preference to partition to extractant is desirable.	Equilibrium partition coefficient of contaminant between soil and flushing solution.
Hydraulic Conductivity	Good conductivity allows efficient delivery of flushing fluid.	Geologic characterization (hydraulic conductivity ranges).
Soil Surface Area	High surface area increases sorption on soil.	Specific surface area of soil.
Carbon Content	Flushing typical more effective with lower soil organic content.	Soil total organic carbon
Soil pH and Buffering Capacity	May affect flushing additives and construction material choice.	Soil pH, Buffering capacity.
CEC and clay content	Increased binding of metals, sorption and inhibit contaminant removal.	CEC, Composition, Texture.
Water Solubility	Soluble compounds can be removed by flushing.	Contaminant solubility.
Soil Sorption	Higher capacity of contaminant to sorb to soil decreases flushing efficiency.	Soil sorption constant.
Liquid Viscosity	Fluids flow through the soil more readily at lower viscosity.	Fluid viscosity at operating temperature.
Liquid density	Dense insoluble organic fluids can be displaced and collected via flushing.	Contaminant density at operating temperature.

Table 4. Design Considerations for Induced Fractures.

Factor	Favorable	Unfavorable
Fracture form	Gently dipping for vertical wells	Vertical (fracture may reach ground surface)
Formation permeability	Moderate to low ($k < 10^{-8}$ cm^{-2})	Unnecessary in high permeability formations (clean sand)
Fracture permeability	>1,000 times formation k	<100 times formation k
Formation type	Rock or fine-grained sediment	Coarse-grained sediment
State of stress	Horizontal > Vertical (over-consolidated)	Horizontal < Vertical (normally consolidated)
Well completion	Access to each fracture most versatile	Screen to several fractures less versatile but less costly than individual access
Site conditions	Open ground over fracture	Structures sensitive to displacement over fracture

type of fluid, rate or pressure of injection, and configuration of the borehole. These factors specifically target relatively shallow applications typical of many contaminated sites. Table 4 shows design consideration.

Summary

In situ remediation of the heavy metals in soils is the reduction, extraction, removal in the subsurface to a level acceptable for site closure. Although many good in situ remediation technologies have been developed, it still demands more than one remediation technology for effective and cost-saving remediation of the contaminated soil. The selection and use of technologies requires to clearly understand the site-specific chemistry of the contaminants and preferred pathways (porosity, permeability and other geologic parameters) within a geologic perspective. Also limiting factors for an intended technology should be critically reviewed to optimize the design of the integrated remediation technology because the integrated technology should treat heavy metals with expectation of benefits over existing ones in many aspects including environmental impacts, cost effectiveness, treatment flexibility, and breadth of applications.

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복합복원기술 개발을 위한 설계안 : 중금속 오염토양을 위한 토양세척과 토양파쇄의 통합 사례 연구

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중금속으로 오염된 농경지 토양을 효과적으로 보권하기 위해서는 토양 매체내에서 반응과정을 거치는 중금속의 동태와 이동성에 따라 한 가지 이상의 복원기술이 선택되어야 한다. 오염토양 복원 시 중요한 토양의 물질적·수리적 요인은 투수성, 공극성, 토성과 토양조직, 오염물질의 형태와 농도, 오염물질의 동태와 이동특성이다. 따라서 중금속으로 오염된 농경지 토양에 적용할 수 있는 복합복원기술 개발 방법을 기존의 사용하고 있는 적용 가능한 화학적 기술과 물리적 기술을 중심으로 검토하여 보았다. 심층토내 중금속을 제거하는 화학적 기술로서 토양세척이 있으나 이러한 단일 기술로는 효과적 복원이 어렵다. 따라서 토양세척기술이 가지고 있는 단점을 보완하기 위한 물리적 기술로 토양파쇄 기술이 있다. 그러나 토양세척과 토양파쇄 기술은 혼용하여 오염토양에 적용할 지라도 오염물질제거율은 높은 이류 흐름 지역에서는 확산유동에 의해 비율제한적으로 된다. 그러므로 선택된 두 가지 기술을 현장에서 효과적으로 적용하기 위해서는 오염토양 복원 시 공정별로 각각의 기술이 가지는 장단점을 파악하기 위해서는 기존 현장에서 기술 적용 시 발견된 문제점과 요인들을 검토하여야 한다. 또한 복원의 효율을 예측하기 위해서는 오염물질의 역학적 탈착, 유동과 이동을 포함하는 예상수학모형을 통해 오염토양의 이질성과 복합 반응에 의한 실질 심층토내에서의 유동 경로 정확한 특성을 파악하여야 한다.