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OFFICE OF TECHNOLOGY DEVELOPMENT INTERGRATED PROGRAM FOR DEVELOPMENT OF IN SITU REMEDIATION TECHNOLOGIES

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OFFICE OF TECHNOLOGY DEVELOPMENT

INTEGRATED PROGRAM FOR DEVELOPMENT OF IN SITU REMEDIATION TECHNOLOGIES

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ABSTRACT

The Department of Energy's Office of Technology Development has instituted an integrated program focused on development of in situ remediation technologies. The development of in situ remediation technologies will focus on five problem groups: buried waste, contaminated soils, contaminated groundwater, containerized wastes and underground detonation sites. The contaminants that will be included in the development program are volatile and non volatile organics, radionuclides, inorganics and highly explosive materials as well as mixtures of these contaminants. The In Situ Remediation Integrated Program (ISR IP) has defined the fiscal year 1993 research and development technology areas for focusing activities, and they are described in this paper. These R&D topical areas include: nonbiological in situ treatment, in situ bioremediation, electrokinetics, and in situ containment.

INTRODUCTION

In June 1991, the Department of Energy's Office of Technology Development instituted an integrated program focused on development of in situ remediation technologies. In situ remediation is defined to include both treatment and containment technologies. In this integrated program, in situ treatment technologies refer to those that will treat contaminated areas without requiring the material's excavation or removal, and also to technologies that will enhance the contaminants' removal from the subsurface. In situ containment technologies refer to those that slow or stop the migration of contaminants to the surrounding environment. Implementation of in situ remediation technologies to clean up DOE sites can:

- minimize health effects on workers and the public by reducing contact exposure
- reduce the costs for cleanup by orders of magnitude by eliminating the need for waste excavation, transport, and disposal
- enable the remediation of areas that currently are not accessible, such as the deep subsurface and areas beneath structures

Advantages are seen in having developed and available several technology options. This is particularly true where state or local regulations may prohibit the use of certain techniques or where the proposed land use favors one technology over another.

The objectives of the In Situ Remediation Integrated Program (ISR IP) are to 1) develop and manage the R&D activities for in situ remediation technologies for hazardous wastes, radioactive wastes and mixed wastes in soils, groundwater, and storage tanks at DOE sites; 2) coordinate the technology development to avoid duplication of effort and to maximize information feedback needed to focus and integrate the R&D program; 3) conduct applied R&D on in situ remediation technologies to develop the technology to the demonstration point, then transfer the technology to users such as the DOE Office of Environmental Restoration and Office of Waste Operations, other federal agencies, and industry; 4) support the advancement of innovative technologies to assess their potential benefit; and 5) provide additional scope to ongoing in situ remediation technology development activities so that additional data can be collected and the technology can be extended to other DOE sites. To accomplish these objectives, the ISR IP has formed teams of end users and technical experts to aid in identifying technology needs, prioritizing R&D activities, and reviewing progress and performance of the technology development activities.

PROBLEM DESCRIPTIONS AND TECHNOLOGY NEEDS

To focus the R&D activities for in situ remediation technologies, the ISR IP has been extracting problem descriptions, technology functional requirements, and technical issues from the Integrated Demonstrations, waste operations, and environmental restoration. In addition, technology development activities within other DOE organizations, the EPA, DOD, and industry are being evaluated for applicability to the ISR IP. The ISR IP will prioritize DOE problem groups and contaminants and then focus the available R&D resources on developing technologies for the high-priority problems. The problem contaminants existing throughout the DOE complex include both volatile and nonvolatile organics such as trichloroethylene (TCE), polychlorinated biphenyls (PCBs), perchloroethylene

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(PCE), carbon tetrachloride (CCl_4), chloroform, gasoline components, and diesel fuel components; radionuclides, including plutonium, uranium, tritium, transuranic elements, radium, thorium, cesium, and strontium; inorganics such as antimony, arsenic, beryllium, cadmium, chromium (VI), lead, mercury, and nitrates; and highly explosive materials. These contaminants are found in a variety of matrices and waste forms, including groundwater and surface water, arid and nonarid soils, and various liquid and solid waste forms.

In situ remediation technologies are applicable to five DOE problem groups: buried waste, contaminated soils, contaminated groundwater, containerized waste, and underground detonation sites. These problem groups, most of which contain mixed low-level wastes, were identified in conjunction with a technology needs assessments performed under the direction of the Office of Environmental Restoration.[1] These problem groups described below, which are amenable to in situ remediation, represent about two-thirds of the known release sites throughout the DOE complex.

Buried Waste

The buried waste category includes landfills, trenches, and other types of burial grounds. They may be unlined and may or may not be capped; they are a current or potential source of vadose zone and groundwater contamination. The burial grounds typically contain heterogeneous contaminants and contaminated media. Burial grounds can be contaminated with low-level radioactive waste and nonhazardous waste, such as animal carcasses, sewage sludge, construction debris, etc. They may also contain mixed radioactive (high-level, low-level, and TRU) and hazardous waste (organic, inorganic, or metals). Wet solids, including sludges, absorbed liquids, resins, and cemented sludges, can have high organic content, along with hazardous metals and radionuclides. Homogeneous dry solids, including grout, concrete, asphalt, brick, soils, salt, and ash, are contaminated with hazardous chemicals, hazardous metals, and/or radionuclides. Large heterogeneous dry solids, including equipment, gloveboxes, glass, construction debris, and metal, are contaminated with hazardous chemicals, hazardous metals, and/or radionuclides. Small heterogeneous dry solids, including filters, glass, laboratory equipment, plant equipment, combustibles, ceramics, glassware, leaded rubber gloves, and lead, are contaminated with hazardous chemicals, hazardous metals, and/or radionuclides. Mercury-contaminated equipment and sludges also exist.

Contaminated Soils

Contaminated soils are found in basins and ponds, around high-use areas associated with existing facilities, and at sites affected by contaminant-specific events. This problem group is described in the following three paragraphs:

Basins and ponds consist of percolation, evaporation, and/or seepage basins and ponds; trenches; sumps; drainfields; and cribs. These formations usually received effluent generated from site activities. They are unlined and pose a current or potential source of vadose zone and groundwater contamination. The basins and ponds can consist of low-level radioactive waste with RCRA metals, such as chromium, lead and mercury, and/or organics, and hazardous waste that includes organic and/or inorganic constituents.

The high-use areas include contaminated soil associated with aboveground or underground storage tanks, piping sewer lines, and buildings. Characterization and remediation are complicated by location of the soil around tanks and piping and beneath buildings. The contaminated soil is often localized and is associated with piping and sewers located in or near areas used daily. Generally the subsurface contamination does not provide an opportunity for direct contact, but presents a current or potential source of groundwater and vadose zone contamination. The soils around piping, tanks, and buildings can be contaminated with radioactive (high-level waste, low-level waste and/or TRU) and hazardous waste (metals, organics, and inorganics).

The general-soils problem category includes soils contaminated by a contaminant-specific episode or event (leak, spill, effluent discharge, or weapons-testing explosion). It provides a current or potential source for vadose zone and groundwater contamination. These soils can contain organics such as PCBs, volatile organic carbons (primarily TCE and carbon tetrachloride), and petroleum-related constituents (sometimes free product in the subsurface). These soils can also be contaminated with mixed low-level radioactive waste and hazardous waste such as organic, inorganic, and metal (chromium, cadmium, lead, and mercury) contaminants, and high explosives. Sediments in ponds, streams, and rivers are contaminated with low-level radioactive waste received through runoff from surrounding areas or from discharge of contaminated groundwater. The potential exists for sediment transport to other surface water bodies used for drinking water and/or recreational purposes.

Contaminated Groundwater

Contaminated groundwater plumes range from several acres to hundreds of square miles, and the extent of contamination is unknown in many cases. Contaminants may be present in perched zones and/or confined or unconfined aquifers, including regional and sole-source aquifers. Contaminant concentrations in most cases exceed standards, and offsite contamination may exist. Groundwater may be contaminated with organic hazardous constituents, predominantly TCE and its associated degradation products, but it also may include petroleum products, CCl_4, and high-explosive compounds. Groundwater is also contaminated with organics, metals, uranium, tritium, and other radiological constituents. The contamination resulted from leaking tanks or piping, surface spills, injection wells, or discharge from disposal sites, and is located several hundred feet below the surface.

Containerized Wastes

Containerized waste consists of sludges in tanks with radioactive waste (high-level waste, low-level waste, and TRU), hazardous waste, and drummed waste. Sludges or similar types of mixed wastes in partially aboveground or underground storage tanks are usually low-level waste forms, but high-level wastes also may be present. The waste may be sludge-like, dust-like, liquid, or solid, and will require removal or stabilization in the tanks prior to decontamination and decommissioning of the tanks. The tank integrity is questionable, and tank leaks present a current or potential source of surface soil, vadose zone, and groundwater contamination.

Underground Detonation Sites

This problem category includes underground weapon or nuclear test sites. The contaminants include low-level radioactive and hazardous (RCRA metals, organics, and/or inorganics) constituents. The contaminated areas are generally isolated from the surface. They represent a current or potential source of groundwater contamination because detonations were above and below the water table.

PROGRAM TECHNICAL AREAS

The problem areas just described led to the identification of specific technical program areas for developing in situ remediation technologies. The ISR IP encompasses three major program areas: treatment, containment, and subsurface manipulation. These major program areas are described in the following sections.

Treatment Technologies

In situ treatment technologies will be developed and evaluated for remediation of volatile and nonvolatile organics, radionuclides, and inorganics in a variety of matrices. The focus will be on technologies associated with in situ destruction, enhanced in situ removal and extraction, and in situ immobilization. Process monitoring and control technologies will be developed for the specific remediation technologies developed under the ISR IP to ensure that technical and regulatory requirements of the end user are satisfied.

In Situ Destruction Technologies. Biological, chemical, and thermal technologies will be reviewed, and the R&D activities will be identified that are required to support demonstration of these technologies for the destruction of organics and nitrates. Special emphasis will be given to treatment of these contaminants in the presence of radionuclides and heavy metals. The goal is to develop technologies that reduce toxicity through destruction of contaminants without adversely impacting the environment. Technologies under consideration include, but are not limited to, in situ bioremediation, in situ chemical oxidation, and in situ electrochemical oxidation.

In Situ Removal and Extraction Technologies. This program area will review physical methods that can be enhanced by thermal, chemical, or biological subsurface treatments to extract contaminants from the subsurface for their destruction or immobilization in aboveground treatment facilities. R&D activities required to support demonstration of these technologies will be identified. Emphasis will be on the extraction of radionuclides from soils and of mixtures of mobile organics and metals from soils. The goal is to develop technologies that reduce toxicity by removing contaminants from the affected environment to meet acceptable levels. Technologies under consideration include, but are not limited to, in situ soil flushing, dynamic steam stripping, in situ heating, and electrokinetics.

In Situ Immobilization Technologies. This program area will focus on 1) thermal or chemical methods that immobilize contaminants and soil matrices in a waste form created in place and 2) biological or chemical methods that can alter redox conditions in soil or groundwater to chemically immobilize the contaminants. The goal is to develop technologies that will reduce the exposure hazard by immobilizing contaminants within the environment. The technologies under consideration include, but are not limited to, in situ precipitation, in situ redox manipulation, in situ solidification, and in situ grouting.

Containment Technologies

This program area will evaluate physical techniques that will isolate or contain contaminants in a defined zone prior to treatment and/or during treatment. The focus will be on four areas: 1) the remediation of heterogeneous waste sites such as landfills, inactive burial trenches, and surplus facilities; 2) the closure of engineered waste systems such as grout vaults and active burial trenches; 3) interim containment of "leaking structures"; and 4) the surface control of contaminant dispersal during in situ remediation. The goal is to develop physical systems to prevent offsite dispersal of contaminants into the environment through air, surface water, or groundwater pathways. The technologies under consideration include, but are not limited to, hydraulic isolation, cryogenics, in situ grouting technologies, vitrified subsurface barriers, and the permanent isolation barrier system. Process monitoring and control technologies will be developed for the specific remediation technologies developed under the ISR IP to ensure that the end user's technical and regulatory requirements are satisfied.

Subsurface Manipulation

This program area will evaluate physical systems that can be used to avoid dispersal of contaminants, in addition to being used for the dispersal of treatment agents during in situ remediation processes. These systems must be compatible with in situ remediation processes. This program area will crosscut the treatment and the containment technologies because many of its physical systems can be used for technologies in each program area. The goals are to develop physical containment systems to ensure that adequate control of the subsurface remediation process is maintained. Technologies under consideration include, but are not limited to, electrokinetic migration of contaminants, hydraulic isolation, auger or jet mixing, hydro- or cryofracturing, pneumatic fracturing, and vacuum-vaporizer well systems.

PROGRAM INTERFACES

The ISR IP is emphasizing primarily the establishment of interfaces and the coordination to develop and demonstrate ISR technologies. The ISR IP has established interfaces with the fundamental R&D organizations and the clients. Figure 1 depicts the interfaces and clients. Basic R&D provides for the advancement of fundamental knowledge and the discovery of new concepts and principles. The ISR IP will link with the fundamental R&D organizations to promote the transfer of knowledge for focusing applied R&D activities and extending the application of the technology. Technical issues requiring fundamental R&D efforts will be identified by the ISR IP and discussed with the appropriate organization for resolution. The ISR IP will also interact with other organizations and/or programs that are conducting applied R&D to minimize duplication of effort and identify technology gaps. The ISR IP has three primary clients: the Integrated Demonstrations, the Office of Environmental Restoration, and the Office of Waste Operations. Problem descriptions and technology functional requirements will be identified for each client and used to determine the program areas for initiating R&D activities. The ISR IP will provide the clients with developed technologies for demonstration.

FISCAL YEAR 1993 R&D TECHNOLOGY AREAS

Technical support groups, consisting of experts from DOE contractors, other federal agencies, universities, and private business, have defined the potential promising areas for DOE to fund technology development for in situ remediation technologies. Four groups convened and categorized research and development needs into the following areas: in situ bioremediation technologies, in situ electrokinetics remediation technologies, nonbiological in situ treatment technologies and in situ containment technologies. Each group defined specifications for the promising research within these different areas and prioritized those areas within their groups. The ISR IP is now seeking R&D proposals that will address these R&D technology needs. These needs are described in more detail below:

<u>Nonbiological In Situ Treatment</u>

Nonbiological In Situ Treatment R&D Areas focus on the following:

- In situ destruction and treatment of contaminants in groundwater and soils. In situ treatment concepts for development include chemical, electrochemical and thermal destruction of TCE, PCE and CCl_4, chemical and electrochemical reduction of chromium, uranium, and nitrate and treatment of radionuclides.

- In situ container destruction as pretreatment for buried waste. A variety of in situ treatment technologies might otherwise be applicable to buried waste that they cannot yet access safely any containerized contaminants. Techniques for destroying most or all ambient container integrity in buried waste need to be developed.

- Subsurface manipulation - addition, dispersion, and recovery of materials in soil for in situ treatment. Contaminants need to be treated in place or removed and recovered for aboveground treatment without disturbing the soil or buried waste. The engineering problems of adding, dispersing (mixing) and recovering treatment materials and residuals without disturbing the soils currently prevent the in situ application of many available and innovative treatment processes. Proposed R&D is sought for solutions to the following: How can treatment materials, nutrients, and flushing/extraction agents be moved into unsaturated and saturated soils? How can contaminants and treatment residuals be removed from soil? How can materials be mixed or brought into uniform contact with soils? How can the effectiveness of in situ mixing be measured in order to distinguish any mixing effects from the effect of the treatment or recovery process? How can the movement of liquid or gaseous treatment materials in unsaturated soils be controlled so that they do not escape from the treatment area? How can reaction products/residuals be separated from soil particles in situ so that they can be removed from the soil?

- Permeable barriers for containment of residual contaminants in groundwater. Permeable treatment barriers which would retain organic compounds or radionuclides while permitting the passage of groundwater could be used to prevent the further migration of the contaminants. Proposed R&D would lead to the selection of sorbents that are nontoxic, not subject to significant degradation, and inexpensive, and then the mechanism for forming the barrier must be developed based on the sorbent's properties and the site's hydrogeology.

- In situ container combustion of buried waste. Most buried waste is actually solid and contains both combustibles and noncombustibles. If most of it could be combusted in situ, toxic and hazardous organics could be destroyed and the bulk volume of combustibles eliminated. Residuals of any hazardous inorganic materials would remain in the burial grounds and could be stabilized in situ and, hopefully, converted in the overall process from a mixed to a radioactive classification. A technology needs to be developed, demonstrated and tested for efficient combustion in the shallow subsurface.

<u>In Situ Bioremediation</u>

The in situ bioremediation R&D needs will focus on the following:

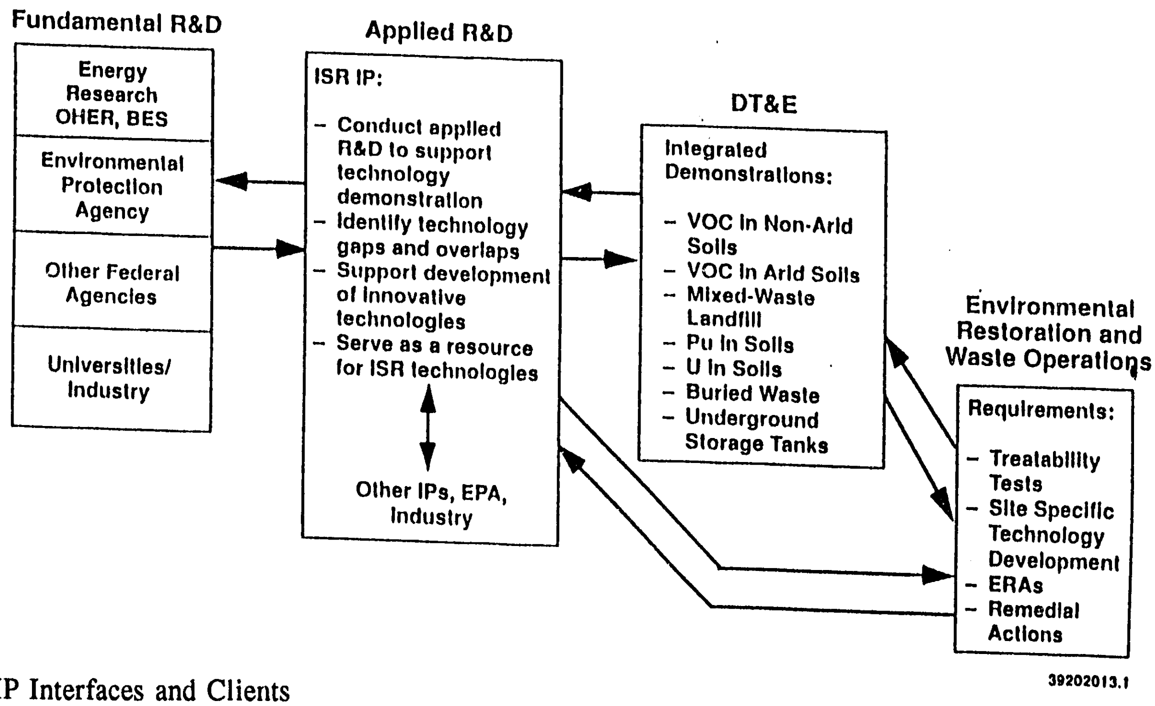

Figure 1. ISR IP Interfaces and Clients

- Techniques to evaluate the extent of bioremediation and the fate of the contaminants. In situ bioremediation of soil and groundwater contaminated with organics and mixed waste can not be applied to DOE sites as part of remediation activities unless the effectiveness of the process can be demonstrated to the regulators. The proposed R&D should result in the development of techniques and tools that can be utilized in an integrated fashion to demonstrate, through simulations monitoring and rigorous statistical techniques, the extent of biodegradation and the fate of contaminants in the targeted treatment zone.

- Bioremediation of mixed wastes. Biotechnology for in situ treatment of mixed wastes can be applied in three areas: 1) bioimmobilization of metals and radionuclides through bioaccumulation, biosorption or biologically assisted oxidation or reduction (biostabilization), 2) bioleaching for mobilization of specific metals and radionuclides, and 3) biological destruction of organics and/or nitrates in the presence of radionuclides/metals. R&D proposals are being sought for these technologies.

- Delivery of nutrients and/or microorganisms to contaminants and the mixing of the three to develop a good in situ environment. Chlorinated and petroleum hydrocarbons are among the most prevalent organic contaminants at DOE sites. These relatively mobile chemicals seep below the ground surface to contaminate the vadose zone and groundwaters. To achieve effective in situ bioremediation, the appropriate microorganisms, the nutrients they need for growth, and the contaminants of concern must be brought together and mixed, and an appropriate environment (pH, temperature, absence of toxicity, etc.) must be created. Proposed R&D is required to develop, evaluate and optimize potential methods for introducing and mixing contaminants, microorganisms, and nutrients into groundwater, the vadose zone, and soils for enhancing in situ biodegradation of contaminants.

- Bioremediation of chlorinated solvents. Improved technologies are also needed for 1) coupling of anaerobic and aerobic processes in situ, 2) treatment of mixtures of TCE, PCE, $CHCl_3$, and CCl_4, and 3) treatment of these solvents that are retained in the vadose zone.

- Development of standardized treatability studies. Methods, procedures and protocols need to be developed, modified, compared and evaluated for treatability of different wastes using in situ bioremediation techniques.

- Bioremediation of nonaqueous phase liquids (NAPLs). At DOE sites, the presence of pure organic product or NAPLs in the subsurface has been confirmed. Technologies are needed to effectively destroy the NAPLs in the subsurface or enhance their migration or dissolution to improve the potential for subsequent degradation or recovery.

Electrokinetics

The R&D needs for electrokinetics were based on the results of a DOE-sponsored workshop on electrokinetics. FY 93 electrokinetics R&D activities will focus on applying the technology to soils contaminated with heavy metals and radionuclides. The R&D will include 1) treatability tests using arid soils, 2) treatability tests using humid soils, and 3) process development to gain an understanding of the technology in order to evaluate its application at DOE sites. Process development activities would support the treatability tests and would provide: a scientific explanation for the migration and removal of heavy metals and radionuclides in both arid and humid soils, the effects of soil heterogeneity on contaminant migration, effects of contaminant speciation, and the controlling parameters for contaminant migration; a system for the removal of contaminants within the soil and at the electrodes; and the engineering design data required for scale-up of the technology.

In Situ Containment

In situ containment R&D areas focus on five areas:

- Hydraulic and diffusion barriers in the vadose zone surrounding waste areas. Buried waste and contaminated soils in the vadose zone are subject to several mechanisms that can mobilize contaminants and conduct them to the groundwater or to the surface. Caps installed over waste areas may be effective in minimizing the amount of atmospheric water that advects into and through the waste area; however, they may not be effective in limiting the amount of contaminants that may migrate horizontally from the waste unless the caps extend significantly beyond the waste zone's boundary. If a vertical diffusion/hydraulic barrier could be constructed in the vadose zone as a ring around the waste area, the required area of the cap could be substantially reduced and the potential successful performance of the cap could be increased.

- Asphalt technology for design of infiltration barriers. Protective surface barriers designs will require a systems approach. A method to ensure optimum and extended performance of the barrier is to construct a subsurface layer using asphalt. The effectiveness of asphalt in limiting water movement to extremely low levels needs to be evaluated.

- Physical and chemical fixation of surface contamination. In arid regions, surface and near-surface contamination can be spread through various mechanisms such as air suspension as dust and aerosols, animal transport, and vehicular agitation during restoration activities. Physical and chemical fixation technologies are needed to prevent further dispersion of contamination pending the completion of necessary remedial actions. Proposed R&D is sought on the novel application of commercially available fixants and on the development of fixants for specific DOE contaminants.

- Subsidence control for buried waste. Most buried waste sites throughout the DOE complex contain degradable wastes (e.g., wood, paper, carbon steel drums, plastic containers, rubber, etc.) well as large voids within and among waste components. The physical degradation of buried wastes, the consolidation of earthen backfill, and the sedimentation of soil within voids cause the cover to subside (or settle). This settling compromises the integrity of the cover, creating surface depressions and greatly increasing the chances for water infiltration. There is a need to physically stabilize buried waste sites to ensure against cover failure and provide long-term protection against water and biointrusion. Proposed R&D is sought for the demonstration of subsidence control technologies at an actual or surrogate waste site.

- Physical, chemical or biological control of contaminant transport in the vadose zone. Much of DOE's wastes and/or contaminant plume "source terms" are associated with the vadose zone. There is a need to develop technologies that will effectively contain the existing plumes and control source terms. The technical concept is to emplace a permeable physical, chemical or biological barrier into the vadose zone below the source location to attenuate, retard or remove the contamination from the transmitted groundwater. R&D proposals for developing these barriers are sought.

CONCLUSIONS

The ISR IP has defined its objective, scope and major program areas. Fiscal year 1993 research and development areas for investment have been defined and proposals are being solicited. Completion of these research and development activities will result in the advancement of nonbiological in situ treatment, in situ bioremediation, electrokinetics and in situ containment technologies.

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