SEPARATIONS TECHNOLOGY ROADMAP FOR DOE ENVIRONMENTAL MANAGEMENT

Sharon Robinson, Jack S. Watson
Oak Ridge National Laboratory
PO Box 2008, Oak Ridge, TN 37831

Jerry Harness
Oak Ridge Operation Office, U.S. Department of Energy

Dan Melamide
U.S. Department of Energy/EM, Germantown, MD

Ian Tasker
Concurrent Technologies Corporation

ABSTRACT

Chemical and physical separations are critical to the Department of Energy's (DOE's) long-term efforts to cleanup in a cost-effective manner the environmental contamination and accumulated wastes in the nation's nuclear weapons complex and are incorporated into many baseline and alternate approaches. The purpose of this roadmap exercise was to provide a cohesive examination of the requirement for near- and long-term separations science and technology research to support implementation of baseline flow sheets for all sites as well as longer-term efforts to develop alternate flow sheets for the high-cost and/or high-risk projects. Alternatives that could provide major cost savings and acceleration of cleanup/closure schedules were also addressed. R&D efforts that existing at the close of FY2001 were examined to evaluate any gaps between the needs and the existing programs as well as opportunities that could result in major improvements in performance, costs, and/or schedules. The roadmap was assembled by the staff of the Efficient Separations and Processing Crosscutting Program, but it addressed needs and separations activities that were being pursued in other programs, including industrial contractors and university studies. Information for the roadmap was obtained from DOE-EM organizations at all major sites, from focus areas, and from scientists who understood both the site needs and the potential technologies that could have a major impact in improving site and waste cleanup.

INTRODUCTION

Although there has been a general understanding that separation technologies are critical to the effective and economical closure of DOE-EM facilities, there had been no unified examination or strategic planning for separations R&D within the EM complex. The importance of separations was stated particularly clearly (1) in the DOE report in 2000 on *Adequacy Analysis of the Environmental Quality Research and Development Portfolio.*

> Separations are essential to environmental quality for the purpose of isolating toxic substance from non-toxic media to allow the former to be economically managed. For example, managing the large volumes of tank waste and contaminated soils and groundwater without separation of toxic constituents would not be economically feasible. In theory, all problem areas involve separation of toxic substances from solid media (facilities, equipment, soils), liquids (groundwater), and stored waste (tank wastes, mixed wastes, spent fuel). Separations technologies are crosscutting because they are used in so many DOE programs and applications. The essence of the crosscutting problem is economic, efficient separation of very dilute toxic substances from not-toxic media while minimizing the amount of the latter accompanying the former.

To perform a unified evaluation of the separation needs and opportunities, a roadmap activity was organized by the Efficient Separations and Processing Crosscutting Program (ESP-CP) but incorporated input from those leading EM activities at the major sites and from key research personnel, including leaders of the focus areas and important research scientists and engineers. The roadmap process included face-to-face discussions with each when possible. There were follow-up phone discussions and a major workshop with more that 40 key people participating in interactive discussions on separations needs, opportunities, and gaps in the current (FY2001) programs. The information from this workshop and the numerous discussions were assembled into a draft roadmap document by the staff of the ESP-CP. That draft was then distributed all sites, all focus areas, and all individual participants for comments and suggested changes. This special effort was done to make the resulting roadmap a truly DOE-wide document, not just opinions of the ESP-CP staff. The evaluations considered separation needs being addressed by numerous programs within and (to the extent possible) outside DOE. Only a small portion of the total separations R&D considered was sponsored at that time by the ESP-CP.

ORGANIZING R&D NEEDS

Although the needs were collected from specific sites and focus areas, the crosscutting nature of separations technologies and applications called for a different grouping of the needs and technologies. The wastes were divided into nine groups according to their properties.

> Dilute aqueous wastes
> Concentrated aqueous wastes
> High-activity solids
> Soils and sediments
> Combustible solid wastes
> Contaminated metal, debris, concrete, and other non-combustible solids
> Gas streams
> Tritium

Unique wastes

Each of these wastes involves different problems and opportunities and was more effectively evaluated separately.

Needs and opportunities were evaluated in terms of three time frames, near-term (less than 10 years), immediate-term (10 to 20years), and long-term (20 to 30 years). For each waste group and each time period, the needs were placed in a priority that identified them as of high-, medium-, or low-priority. The priorities reflect both the risk of baseline technologies and the potential benefits of R&D on alternate technologies. By necessity, most current R&D in EM is directed at near-term needs. This reflects the need for immediate decision in some cases and the need to start construction of facilities to meet schedules in other cases. However, it because apparent during the roadmap exercise that some wastes will not be treated for several years and that some facilities will be operating for 20 years or more and will need to be repaired and upgraded during the next 20 to 30 years. The "cut-off" of the study at 30 years reflects both the optimism that EM will be able to solve most, if not all, of its major waste problems in this time frame and the recognition that technology predictions beyond 30 years seldom have increasing uncertainty.

KEY FINDINGS

All of the DOE sites have near- to mid-term needs for separations-related data, technology development, and baseline technology performance verification. Such needs were, for the most part, being addressed by existing EM research program. However, additional R&D could lead to significantly improved flow sheets and, thus, to significant cost savings.

Additional uses of separations that can have a particularly large impact on cost and schedule are related to the following types of remediation activities. These include:

- **Reduction or Stabilization of High-Activity Waste**

  Removal of sodium salts, organics, and metals from radioactive alkaline sludges to reduce the volume of vitrified high-level wastes; treatment of high-aluminum content acidic calcine to avoid direct vitrification and disposal as high-activity wastes; and removal/stabilization of radioactive components in sludge heels, which impact closure.

- **Environmental Restoration**

  Development of highly selective sorption materials for removing toxic organics and metals from soils and groundwater; methods to fix or remove these contaminants from loaded sorption materials; and fundamental understanding of the effects of separations chemistry on pollutant transport for use in technology design and risk evaluations.

- **Decontamination and Decommissioning**

  Separations of radionuclides and metals from metal, debris, and concrete to reduce the volume and/or lower the regulatory category of the waste sent to disposal.

Because of the high cost and long schedules associate with remediation of DOE sites, there is also a potential for significant impacts from long-term science and technology development. The FY2001 portfolio of projects were deficient in addressing needs at the applied research and development stages. It consisted of 57 basic science projects, 4 applied research projects, 13 exploratory and advanced development projects, and 46 engineering development and deployment projects. Scientific research and applied technology activities focused on longer-term, high-risk, and high-cost portions of the flow sheets could lead to significant improvement that could be implemented during future plant upgrades. These improvements would reduce waste generation (and disposal), operational risks, and cleanup time.

Lack of backup technology development is a potential barrier to achieving crucial EM goals. Sites require backups or alternatives to baseline technologies to baseline flow sheets in cases where unforeseen technical or regulatory problems are most likely to occur. The importance of having such a backup technology was illustrated when an alternative process to In-Tank Precipitation (ITP) was needed for removal of cesium from tank waste at SRS. Before problems with ITP were evident, separations research by the ESP-CP and the EMSP had identified two technologies (ion exchange with crystalline silicotitanates and a solvent extraction process) that were potential alternatives to ITP. Similar backup technologies are needed for other high-profile and high-cost baseline projects, including high-level waste treatment at Hanford and Idaho. Recent pressure on DOE to eliminate incinerators has also created a need for alternate technologies.

Development of cost-effective technologies for decontamination of metals and other solids is severely hampered by the lack of federal regulations governing release or recycle of these materials. Development of such regulations could significantly impact the amount of waste requiring treatment and the separations research needed to support this area.

SPECIFIC FINDINGS

To better understand the finding of the study, one should consult the full roadmap document (2) that explains findings, needs and opportunities. However, a summary of selected highest priority needs and opportunities are presented in Table 1. To simplify the table, all needs are grouped as near-term or long-term, but the full document provides a better description of more needs and the time-scale of those needs. It gives a more precise breakdown of the time scale for needs, a listing more medium as well as high priority needs, and more details on all findings.

Dilute Aqueous Solutions

Contaminant-specific natural biotreatment and sorption systems are needed to extend the life-time and effectiveness of in situ and ex situ barriers. Methods for removing contaminants or for permanent fixation of contaminants within in situ barriers are likely to be important to the long-term acceptance of these technologies.

Concentrated Aqueous Solutions

New separation methods for removing cesium, strontium, and transuranic elements from alkaline and acid wastes are needed as alternates and/or improvements over current baseline flow sheets. Significant technical risk remains in the current baseline flow sheets. Simpler, but possibly less efficient, flow sheets are likely to help in the current efforts to allow some tank wastes to bypass the main waste treatment facilities (baseline facilities) and be sent to less expensive disposal options. Flow sheet optimization could further reduce waste glass productions.

High-Activity Solids

Separation of key radionuclides such as technetium and neptunium from tank heels is needed to enhance approval for tank closure. Removal of metals (chromium, aluminum, and sodium) and sulfate from vitrifier feed can enhance glass waste loading. Drastic flow sheet changes such as acid dissolution of tank sludge followed by a single integrated solvent extraction step (such as the UNEX process) could result in greatly reduced glass production.

Soils and Sediments

Contaminant specific treatment methods are needed that consume less reagents and leach or destroy less soil. These methods would be of particular interest for slightly contaminated soils where it is necessary to remove only a small quantity of contaminant and where very large volumes of soils are involved. This includes improved soil washing reagents and techniques. Removal of transuranium elements from sludges and soils are of particular interest. Fixation by permanent sorption or other approaches is an alternative to contaminant removal, and reliable permanent fixation could allow more contaminants to be left "in-place."

Combustible Solid Waste

Alternative processes are needed to destroy organic contaminants, especially when incineration is unacceptable. Separation methods are needed for multi-regulated wastes, especially to separate radioactive components from organic wastes.

Contaminated Metal, Debris, Concrete, and Other Non Noncombustible Solids

Separation methods are needed to recycle metals such as nickel and to remove radioactivity from bulk metals, other decommissioning wastes, and used equipment. Classified solids present special treatment problems that can involve destruction ("chop and dispose") and/or other processing of components. Equipment used in high-level chemical processing facilities presents especially difficult decontamination and disposal problems; such problems exist currently with old fuel reprocessing facilities and will be occur again in a few decades as new high-level waste treatment facilities are to be decommissioned.

Gas Streams

The most important gas streams to be handled by EM are the off-gas systems from vitrifiers and other waste processing equipment. These gas treatment systems are usually designed as part of the vitrifier or other facilities.

Tritium

Tritium offers especially difficult challenges since there are no efficient isotope separations that can concentrate tritium by large factors in a single stage, as can be done with selective adsorption or ion exchange processes. This means that multiple stage systems with significant reflux are required. One should not expect a simple and inexpensive process to remove trace concentrations of tritium and concentrate the tritium to high concentrations for fixation and disposal. Nevertheless, greatly improved isotope separation methods are possible and could extend the application of tritium removal to more waste streams.

Special/Unique Waste

There are numerous small quantities of unique wastes in DOE facilities that are difficult to handle and dispose by conventional methods. Special separation and/or processing methods may be needed to prepare each of these for disposal. Although the quantities of these wastes may not justify extended R&D efforts, serious thought should be given to each of these problems, and significant R&D on methods for handling several of these wastes may be justified.

Other Findings

In addition to the specific needs identified and discussed, it became evident that there were areas where better understanding of fundamental properties of waste streams or separation methods were needed, especially to reduce the risk that the baseline technologies will fail or to better define the requirements of separation systems. One notable example was the need to better understand the role of adsorption and ion exchange processes in the transport of pollutants through soils (both in the vados zone and in the saturated region). Adsorption and ion exchange are common separations methods, but they also play important roles in contaminant transport in the environment; so the phenomena are also important in assessing stewardship and contaminant release issues.

Another group of separation issues and opportunities were identified that can be called "system interface" issues. These may not appear obvious without a careful examination of both the waste treatment methods and the processes that generate wastes. Facility operations that minimize the production of wastes constitute one set of such issues. Although EM is usually concerned with wastes that were produced in previous production operations, the waste treatment operations themselves also produce waste streams. Some of the key opportunities are concerned with the treatment of high-level tank waste (mentioned above and listed in Table 1) also involve "interface."

For instance, selective blending of wastes from different tanks with different concentrations of key components that affect waste loading in glass could alter the need and requirements for separation processes. Minimization of gas production, and thus secondary wastes produced from the off-gas treatment systems, can be important. The interface between pretreatment (mostly separations) and vitrification of tanks waste is particularly important since the performance of the pretreatment facilities (removal of key components) and the requirement of the vitrification plant to minimize waste production must match.

Improved use of recycle streams to minimize waste production can be important in decontamination operations as well as in treatment of the more complex tank wastes. Because of the size of many old DOE facilities, the volume of waste produced during decommissioning will be exceedingly large. Most of this material will not be contaminated, but all or much of the material may have to undergo some degree of decontamination if it is to go to low cost landfills.

Interface issues with gas treatment systems deserve mention. As noted, these are separation facilities that are usually designed as part of a vitrification or other larger facility. This is not an inappropriate way to design off-gas facilities since they are then designed by those who should be most familiar with source of the off-gas. However, when the waste stream from the off-gas facility is returned to another facility, there is a potential interface issue. The return of silica containing off-gas treatment waste to the waste tanks at SRS produced serious problems.

GENERAL FINDINGS

Most studies that look at R&D needs are likely to find some gaps in current program and needs for additional work, and that was also the case in this study. However, there were also several other interesting general findings. One general finding of this study was the many places and ways in which separation and processing occurs in throughout the baseline EM plans, and there are many places where there are key technical risks in the separation steps. Separation and processing R&D has also been bundled to become part of other facilities (such as off-gas systems for vitrifiers), and that was often the most efficient way to handle those developments. In this manner, a relatively large portion of the EM program has been in some way appropriately devoted to separation and processing issues.

The treatment of high-level waste (tank waste) is potentially one of the most costly items in the EM program, and this is an area where separations play especially important roles. Since the key to reducing the large cost of treating high-level wastes is to reduce the volume of waste that must eventually be vitrified and sent to the repository, the two basic approaches available are to divert as much waste as possible (under regulations) from the high-level waste vitrifiers and for the remaining waste to concentrate (that is, separate) the radioactive components that must go to the repository. To a lesser degree, concentration of contaminants can be one key to reducing the cost of disposing of numerous other waste streams. Thus, separations are potentially key to reducing the ultimate cost of waste disposal for EM. However, the separation approaches used must also be cost effective, and this means that better methods are frequently needed.

It was somewhat surprising to find that both sites and focus area representatives usually agreed that intermediate- and long-term R&D will be needed to reduce or hold down EM costs. The need for immediate work to complete committed facilities is obvious. However, the need to insure that those facilities will operate as planned is less obvious, except to those closest to the technologies. There are numerous sludges, soils, and tank wastes to be treated, and some of these have not been fully characterized. The effectiveness of the planned treatment facilities to handle this range of wastes has often not been demonstrated. Some times even trace components in a waste will alter the performance significantly. The often-mentioned problems with In-Tank-Precipitation at SRS is probably the best known example of this kind of problem. The presence of very high sulfate in some Hanford tanks is a known problem for the baseline vitrifiers. The long-term stability of in situ barriers for groundwater was mentioned earlier.

Finally, there seemed to be an understanding that processing facilities will not necessarily operate for 20 years of more without repair or even modifications. As typical industrial facilities are usually upgraded and improved over such a time frame, there will be opportunities to upgrade the EM waste treatment facilities, and improved separations are likely to be an important part of such upgrades. There are good reasons to believe that we can continue to improve the performance of EM facilities throughout the coming years.

Table 1
Highest Priority Separation Needs and Opportunities

Waste Type	Sub-type	Near-Term	Long-Term
Dilute Aqueous	In situ	Contaminant specific biotreatment	Contaminant specific methods for large volume wastes
		Separations for real-time GW characterization	
	Ex situ	Methods for dilute colloids	Contaminant specific methods for large volume wastes
Concentrated Aqueous	Tank Wastes	Removal of nitrate, sulfate and hydroxide for baseline and other alkaline systems	Alternate acid flow sheet for greatly reducing glass (waste) volume
High Activity Solids	Tank sludge		Removal of organics, mercury, and metals to improve vitrification
Soils and Sediments	In situ	Highly selective methods for target contaminants	Highly selective methods for target contaminants
	Ex situ	Highly selective methods for target contaminants	Highly selective methods for target contaminants
Combustible Solids		Desorption processes for key contaminants	Desorption processes for key contaminants
		Reductive dechlorination of mixed wastes	Reductive dechlorination of mixed wastes
Non-Combustible Solids		Methods for retrieved buried wastes	Methods for retrieved buried wastes
		Methods to decon HLW treatment equipment	Methods to decon HLW treatment equipment

REFERENCES

1. *Adequacy Analysis of the Environmental Quality Research and development Portfolio.* U.S. DOE Sept, 2000.
2. Harness, Jerry, Sharon Robinson, Ian Tasker, and Jack Watson. *Separations Paths to the Future: Technology Roadmap for the DOE Environmental Management.* March 2002.